The European Physical Journal E5, 583-588 (2001)

Hydrodynamics of nematic ferrofluids

E. Jarkova[1], H. Pleiner[1a], H.-W. Müller[1], A. Fink[2], and H.R. Brand[2]

[1] Max-Planck-Institute for Polymer Research, POBox 3148, 55021 Mainz, Germany;
[2] Theoret. Physik III, Universität Bayreuth, 95440 Bayreuth, Germany;

Received: August 29, 2001

Abstract. We derive hydrodynamic equations for nematic ferrofluids (ferronematics) in the limit that the magnetic degree of freedom has relaxed to its equilibrium value. We concentrate on novel dynamic effects linear in the magnetic field. We show that flow alignment, heat conduction, diffusion, thermodiffusion, viscosity and director reorientation are all modified by the presence of an external field. In particular, the new effects describe reversible (irreversible) couplings, where the conventional effects are irreversible (reversible). We discuss, how these effects can be measured. In principle, this description is applicable to conventional nematics, too, although huge magnetic fields are expected to be necessary for detecting the new effects in this case.

PACS. 61.30.-v Liquid crystals – 75.50.Mm Magnetic liquids – 05.70.Ln Nonequilibrium irreversible thermodynamics

1 Introduction

The synthesis of nematic liquid crystals with single-domain ferro- or ferrimagnetic grains, usually denoted as ferronematics, is of great interest for potential applications but also under the scope of fundamental research. With the pioneering work of Brochard and de Gennes [1] the idea came up to intensify the ponderomotive response of a liquid crystal by doping it with a small amount of ferromagnetic particles. Owing to a strong orientational coupling between the magnetic grains and the surrounding nematogen matrix, the susceptibility of the director dynamics was expected to be appreciably enhanced. Indeed, the magnetic field strength necessary to affect the director was predicted to decrease by several orders of magnitude giving control over the orientational state of the liquid crystal by magnetic fields as weak as 100 Oe.

During recent years considerable efforts were undertaken in the preparation of various dispersions of ferromagnetic particles in liquid crystals. Starting with the first report in 1970 of mixing magnetic grains with the nematic phase of MBBA [2], there was a number of reports on the production of mixtures of rod-like and disk-like nematics with magnetic grains [2–5]. In ref. [6] collective magnetic and orientational effects, but no spontaneous magnetization were described for a suspension of $\gamma-Fe_2O_3$ particles in the nematic phase of MBBA. In many systems investigated there were problems with chemical stability. Recently, however, the preparation of stable ferronematic systems has attracted increasing attention [7–15].

In their original work Brochard and de Gennes started from the so-called "rigid anchoring" approximation, implying that the directions of the director $\mathbf{n}$ and the local magnetization $\mathbf{M}$ are perfectly co-aligned. However, with the synthesis of thermotropic ferronematics [6] it became evident that the rigid-anchoring approximation might not be generally applicable. Within the framework of a microscopic model of rod-like ferromagnetic grains Burilov and Raikher [16] reconsidered the surface interaction between the liquid crystalline nematogens and derived an expression for the free energy of a ferronematic. Thereby the orientations of $\mathbf{n}$ and $\mathbf{M}$ were treated as separate degrees of freedom. As in Ref. [1] the strength of the magnetization was assumed to be in saturation, even without external fields. However, the existence of a remanence in ferronematics, being characteristic for a ferromagnetic ordering (spontaneous equilibrium-magnetization in the absence of any external magnetic field) seems to be experimentally unproven yet. Rather the existing substances all seem to be superparamagnetic like ordinary ferrofluids.

In a recent analysis [17] this question has been addressed within the scope of a Landau theory derived from the underlying symmetries of the problem. Based on a Landau-deGennes expansion of the free energy an expression for the free energy was constructed, which entails nematic as well as magnetic contributions. Depending on the signs and magnitude of the magnetic-nematic cross-couplings there is the possibility to have either a direct transition from the isotropic unmagnetized fluid to

[a] e-mail: pleiner@mpip-mainz.mpg.de

a ferromagnetic-nematic phase (spontaneous magnetization) or a two-step transition via a superparamagnetic-nematic state. Interestingly, a phase with spontaneous magnetization but without nematic ordering is not possible within the scope of this model.

Strictly speaking, owing to the admixture of magnetic particles to a nematic both, the nematic and the magnetic degree of freedom have to be taken into account. However, we will restrict ourselves here to situations where the magnetization has relaxed to its equilibrium value parallel to the applied magnetic field. It is then a function of all state variables (and the external field), but has no independent dynamics, i.e. magnetic relaxation effects are disregarded here. This is a good approximation for times slower than typical magnetic relaxation times ($10^{-6}s$). Since the nematic director dynamics is much slower, it is appropriate to keep explicitly the latter (this has nothing to do with the "rigid anchoring" approximation mentioned above, since $\boldsymbol{n}$ can have any orientation relative to $\boldsymbol{M}$). This description is, thus, in principle also valid for ordinary nematics, where no magnetic degree of freedom is present. However, the new effects described here probably require huge magnetic fields to be observable in ordinary nematics, while in ferronematics their detection should be much easier. In ordinary nematics the diamagnetic interaction energy adding up to the director's molecular field is usually considered to be the only relevant magnetic contribution. However, in ferronematics other effects disregarded so far mostly (except in [18]) might become significant. In the following we will concentrate on those effects, which are *linear* in the magnetic field strength H. As this quantity is of negative parity under time reversal such effects can arise in the dynamics only and a variety of new Onsager couplings will appear. These new couplings change the character of the contributions in the field free case from reactive to dissipative or vice versa. The implications of these new couplings will be illustrated by means of a series of different examples.

2 Nematodynamics without magnetic field

Nematic liquid crystals are characterized by an extended set of hydrodynamic variables that comprise those of a simple liquid (density ρ, momentum density $\rho\boldsymbol{v}$ related to the velocity $\boldsymbol{v}$, and entropy density σ, or equivalently free energy density f) and in addition the director $\boldsymbol{n}$, the symmetry variable denoting orientational changes ($\boldsymbol{n}^2 = 1$) of the preferred direction and, in the case of mixtures, the concentration c. In our case the latter is the concentration of the ferromagnetic particles. The hydrodynamic equations are [19–21]

$$\left(\frac{\partial}{\partial t} + v_i \nabla_i\right) \rho + \rho\, div\, \boldsymbol{v} = 0 \tag{1}$$

$$\rho \left(\frac{\partial}{\partial t} + v_j \nabla_j\right) v_i + \nabla_j \sigma_{ij} = 0 \tag{2}$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j\right) n_i + Y_i = 0 \tag{3}$$

$$\rho \left(\frac{\partial}{\partial t} + v_j \nabla_j\right) c + div\, \boldsymbol{j}^c = 0 \tag{4}$$

$$\left(\frac{\partial}{\partial t} + v_i \nabla_i\right) \sigma + \sigma div\, \boldsymbol{v} + div\, \boldsymbol{j}^\sigma = \frac{R}{T} \tag{5}$$

Generally the densities of the currents for heat $\boldsymbol{j}^\sigma$, concentration $\boldsymbol{j}^c$, and momentum, the stress tensor σ_{ij} and the quasi-current Y_i are the sum of two parts, a reversible and an irreversible one, where the former (latter) has the same (opposite) time reversal symmetry as the rest of the equation and leads to zero (positive) entropy production, i.e. it is reactive (dissipative). Within irreversible thermodynamics the dissipative parts can be derived from the dissipation function R (the source term in (5)) as a potential, while the reversible ones do not follow from any potential [22]. The currents read

$$\sigma_{ij} = p\delta_{ij} + \Phi_{lj} \nabla_i n_l - \frac{1}{2}\lambda_{kji} h_k - \nu_{ijkl} \nabla_l v_k \tag{6}$$

$$Y_i = -\frac{1}{2}\lambda_{ijk} \nabla_j v_k + \frac{1}{\gamma_1}\delta^\perp_{ij} h_j \tag{7}$$

$$j^\sigma_i = -\kappa_{ij} \nabla_j T - D^T_{ij} \nabla_j \mu_c \tag{8}$$

$$j^c_i = -D_{ij} \nabla_j \mu_c - D^T_{ij} \nabla_j T \tag{9}$$

with the transverse Kronecker symbol $\delta^\perp_{ij} \equiv \delta_{ij} - n_i n_j$. The conjugate quantities temperature $T = T(\rho, \sigma, c)$, pressure $p = p(\rho, \sigma, c)$, relative chemical potential $\mu_c(\rho, \sigma, c)$, and the 'molecular fields' $h_i(\rho, \sigma, c)$ and $\Phi_{ij}(\rho, \sigma, c)$ follow from the free energy density f (see Appendix (A.1)). This part of the equations, which constitutes the statics of the system, is derived completely independently from the dynamics, eqs.(6-9).

The heat conduction tensor κ_{ij}, the diffusion tensor D_{ij} as well as thermodiffusion tensor D^T_{ij} (related to the Soret/Dufour effects) are symmetric and have the following form containing together six coefficients (thermal conductivity, diffusivity and Soret/Dufour),

$$\kappa_{ij} = \kappa_\perp \delta^\perp_{ij} + \kappa_\parallel n_i n_j \tag{10}$$

The tensor λ_{ijk}, describing flow alignment in (7) and back flow in (6) contains one phenomenological parameter

$$\lambda_{ijk} = (\lambda - 1)\delta^\perp_{ij} n_k + (\lambda + 1)\delta^\perp_{ik} n_j \tag{11}$$

which is reversible, since h_i, Y_i and σ_{ij} all do not change sign under time reversal.

The fourth rank viscosity tensor contains five viscosities

$$\begin{aligned}\nu_{ijkl} = \nu_2(\delta_{jl}\delta_{ik} + \delta_{il}\delta_{jk})\\ +2(\nu_1+\nu_2-2\nu_3)n_in_jn_kn_l\\ +(\nu_3-\nu_2)(n_jn_l\delta_{ik}+n_jn_k\delta_{il}+n_in_k\delta_{jl}+n_in_l\delta_{jk})\\ +(\nu_4-\nu_2)\delta_{ij}\delta_{kl}\\ +(\nu_5-\nu_4+\nu_2)(\delta_{ij}n_kn_l+\delta_{kl}n_in_j)\end{aligned} \quad (12)$$

3 Dynamics in a magnetic field

3.1 Linear vs. quadratic field effects

Let's consider nematics in a magnetic field. An external field breaks the rotational symmetry externally and n_i relaxes according to the diamagnetic anisotropy to its equilibrium orientation, which is either parallel or perpendicular to the external field. The appropriate molecular field h_i reads

$$h_i^{(M)} = -\chi_a H_i(\boldsymbol{n}\cdot\boldsymbol{H}) \quad (13)$$

Usually this is the only effect of an external magnetic field that is taken into account when dealing with ordinary nematic liquid crystals. All other effects (a few of them are discussed in [18]) are assumed to be small and are neglected. In ferronematics the response to an external magnetic field is rather enhanced and such hitherto disregarded effects can become important. There is the (rather trivial) effect that all material coefficients (transport parameters and susceptibilities) can depend on H^2. In addition, if $\boldsymbol{n}$ is perpendicular to $\boldsymbol{H}$ in equilibrium, the system is biaxial and the uniaxial tensors in eqs. (10, 11, 12) are of the well-known biaxial form. Furthermore, there are the magnetic forces, which are described by the Maxwell stress $\sigma_{ij}^{(M)} = -\mu_{eq}(H)H_iH_j$ with the equilibrium magnetic susceptibility $\mu_{eq}(H)$ [23] and by a redefinition of the pressure $p \to p-(1/2)H^2$. However, all these effects (including (13)) are quadratic in the external field strength and represent additions to effects already present. In the following we will discuss additional effects that are linear in the field and represent new effects thus bearing a good chance of being observable in ferronematics.

Since a magnetic field changes sign under time reversal, linear effects are possible in the dynamics only, since in the statics all relations are time-reversal symmetric, that is invariant under time reversal. In the dynamics the currents come in two classes, either reversible or irreversible, meaning time-reversal symmetric and antisymmetric, respectively. The introduction of a linear field then toggles between these two possibilities.

3.2 Flow alignment

As a first example we will consider flow alignment in the presence of a magnetic field. The flow alignment tensor (11) can have additions linear in the field

$$\begin{aligned}\lambda_{ijk}^D(H)=&\lambda_1^D\left(\delta_{iq}^\perp\epsilon_{pjq}H_pn_k+\delta_{iq}^\perp\epsilon_{pkq}H_pn_j\right)\\ &+\lambda_2^D\left(\delta_{ik}^\perp H_p\epsilon_{pjq}n_q+\delta_{ij}^\perp H_p\epsilon_{pkq}n_q\right)\\ &+\lambda_3^D\left(H_j\epsilon_{ipk}n_p+H_k\epsilon_{ipj}n_p\right)\\ &+\lambda_4^D\left(H_qn_qn_j\epsilon_{ipk}n_p+H_qn_qn_k\epsilon_{ipj}n_p\right)\\ &+\lambda_5^DH_p\epsilon_{piq}n_qn_jn_k+\lambda_6^DH_p\epsilon_{piq}n_q\delta_{jk}^\perp\end{aligned} \quad (14)$$

Note that these contributions are all dissipative while (11) represents only reversible ones. Adding up in eq.(7) both contributions into $\lambda_{ijk}^{(Y)} \equiv \lambda_{ijk} + \lambda_{ijk}^D(H)$ then the cross coupling term in (6) reads $\lambda_{kji}^{(\sigma)} \equiv \lambda_{kji}+\lambda_{kji}^D(-H) = \lambda_{kji}-\lambda_{kji}^D(H)$ due to Onsager's relation[1] guaranteeing a positive (zero) entropy production due to the field-dependent (-independent) parts.

In the stationary case all currents and quasi currents are zero (disregarding the thermal degree of freedom here)

$$\nabla_i\sigma_{ij} = 0, \quad \text{and} \quad Y_i = 0. \quad (15)$$

The first condition is identically satisfied for constant shear flow and constant field. The second one

$$-\frac{1}{2}\lambda_{ijk}^{(Y)}\nabla_jv_k + \frac{1}{\gamma_1}\delta_{ik}^\perp h_k^{(M)} = 0 \quad (16)$$

reads explicitly

$$\begin{aligned}n_k(\lambda-1)\nabla_iv_k + n_j\nabla_jv_i(\lambda+1) - 2\lambda n_in_jn_k\nabla_jv_k \quad (17)\\ +\lambda_{ijk}^D(H)\nabla_jv_k + \chi_a'H_in_jH_j - \chi_a'n_in_kH_kn_jH_j = 0,\end{aligned}$$

where $\chi_a' = 2\chi_a/\gamma_1$.

We will solve eq.(17) for a particular case, where the new linear field terms (14) become manifest: The external field lies in the shear plane

$$\begin{aligned}n_x &= \sin\theta\\ n_y &= \cos\theta\sin\varphi\\ n_z &= \cos\theta\cos\varphi\\ H_y &= H\sin\psi\\ H_z &= H\cos\psi\end{aligned} \quad (18)$$

with the shear flow $\nabla_zv_y = S$ and with ψ the angle of the magnetic field with the shear gradient.

Without the new terms it is known that the director also lies in the shear plane (i.e. $\theta = 0$) making an angle φ with the direction of the shear gradient, where φ is given by

$$2S\left(\lambda\cos2\varphi+1\right) = \chi_a'H^2\sin\left(2\left(\varphi-\psi\right)\right) \quad (19)$$

[1] With the currents $C_\alpha \equiv \{Y_i\,,\,\sigma_{jk}\}$ the forces are $F_\beta \equiv \{h_i\,,\,-\nabla_jv_k\}$, since the entropy production $R \sim -h_iY_i - v_k\nabla_j\sigma_{kj} \sim -h_iY_i + \sigma_{kj}\nabla_jv_k$. Then, Onsager's relation $D_{\alpha\beta}(H) = \epsilon_\alpha\epsilon_\beta D_{\beta\alpha}(-H)$, with $C_\alpha = D_{\alpha\beta}F_\beta$, leads to the symmetries stated above for the λ-tensor, since $\epsilon_\alpha\epsilon_\beta = -1$ for cross terms connecting variables of different time reversal symmetry.

The new terms (except λ_2^D) force the director out of the shear plane. Taking first λ_1^D as a representative for the other terms (in order to simplify the formulas) we get the unchanged condition (19) for the in-plane orientation φ of the director. There is, however, now an out-of-plane component of the director given by the non-zero angle θ

$$\tan\theta = \frac{-\lambda_1^D SH\sin\varphi\cos(\varphi+\psi)}{(\lambda+1)\,S\cos\varphi+\chi_a' H^2\sin\psi\cos(\psi-\varphi)} \quad (20)$$

This kinetic expulsion out of the shear plane due to the combined action of shear flow and (in-plane) field occurs, even if the (static) diamagnetic anisotropy would favor a director parallel to the field ($\chi_a > 0$), i.e. to be in-plane. Other contributions in (14) will change also the simple expression (19) for the in-plane components of $\boldsymbol{n}$. E.g. the two last contributions in (14) lead to an out-of-plane orientational angle

$$\tan\theta = \frac{\beta\sin^2\varphi}{2(\lambda\sin\varphi+\alpha\cos\varphi)} \quad (21)$$

with $2\alpha = \chi_a' H^2/S$ and $\beta = (\lambda_5^D - \lambda_6^D)H$. Here the in-plane-angle φ is not given by (19) but follows from

$$\frac{\beta^2\sin^3\varphi}{4(\lambda\sin\varphi+\alpha\cos\varphi)^2} = \frac{1+\lambda\cos 2\varphi-\alpha\sin 2\varphi}{(\lambda-1)\sin\varphi+2\alpha\cos\varphi} \quad (22)$$

In (21) and (22) we have simplified the formulas by assuming $\psi = 0$, i.e. the magnetic field parallel to the shear gradient. For λ_3^D and λ_4^D the appropriate formulas are even more bulky and will not be shown here.

3.3 Heat conduction, diffusion and thermodiffusion

As a second example we consider the heat conduction tensor κ_{ij}. There are additions linear in the field of the form

$$\kappa_{ij}^R(H) = \kappa_1^R\epsilon_{ijk}H_k + \kappa_2^R\epsilon_{ijk}n_k n_p H_p + \kappa_3^R\,(\epsilon_{ipq}H_p n_q n_j - \epsilon_{jpq}H_p n_q n_i) \quad (23)$$

These terms are reversible due to their time reversal behavior, while those of (10) are irreversible. The former are antisymmetric $\kappa_{ij}^R(H) = \kappa_{ji}^R(-H) = -\kappa_{ji}^R(H)$ according to the Onsager's relation thus leading to zero entropy production.

The field-free heat conduction tensor (10) leads to a heat current $\boldsymbol{j}^\sigma = (\kappa_\parallel - \kappa_\perp)\,\boldsymbol{n}\,(\boldsymbol{n}\cdot\boldsymbol{\nabla}T) + \kappa_\perp\boldsymbol{\nabla}T$ that lies in the plane of the director and the temperature gradient. The field-dependent terms lead to (reversible) contributions to the heat current that can be perpendicular to both, temperature gradient and field, or to temperature gradient and director, or to field and director. For example, if the field H is along the x- and temperature gradient G along the y-direction, there is a reversible component of the heat current

$$j_z^\sigma = \begin{cases}(\kappa_1^R+\kappa_2^R)HG \\ (\kappa_1^R-\kappa_3^R)HG\end{cases} \quad \text{for} \quad \boldsymbol{n}\parallel\begin{cases}\boldsymbol{H}\\ \boldsymbol{\nabla}T\end{cases} \quad (24)$$

that is orthogonal to the $\boldsymbol{n}/\boldsymbol{\nabla}T$ plane and perpendicular to the field. This effect is quite analogous to the Hall effect related to electric currents. There, similarly the diagonal elements (the analogues to (10)) are dissipative, while the antisymmetric parts (the analogues to (23)) are non-dissipative [24]. Instead of a voltage transverse to the dissipative electric current, eq.(24) leads to a temperature difference transverse to the dissipative heat current. This effect (especially the part related to κ_1^R) is in principle present in any fluid (and called Righi-Leduc effect [18]) and not restricted to ferronematics, but in the latter system chances are much better that it is observable.

Quite analogously to (23) one can introduce 3 new reversible diffusivities $D_{1,2,3}^R$ and the discussion between (23) and (24) can be taken over replacing ($\boldsymbol{j}^\sigma$, T) by ($\boldsymbol{j}^c$, μ_c). Rather similar is also the case of thermal diffusion. There are three reversible thermal diffusivities $D_{1,2,3}^{T,R}$ of the form (23). Since they are antisymmetric w.r.t. interchange of indices, and linear in the field, they automatically fulfill Onsager's relation $D_{ij}^{T,R}(H) = D_{ji}^{T,R}(-H)$. Under an external magnetic field (of strength H) orthogonal to a temperature gradient (of magnitude G) these new contributions give rise to a concentration current (orthogonal to both) given by the r.h.s. of (24), if the κ^R's are replaced by $D^{T,R}$'s. Thus, Hall-like temperature and concentration gradients are generated, which are transverse to the dissipative heat and concentration currents, respectively.

3.4 Viscosity

As the third example we consider field dependent generalizations of the viscosity tensor

$$\begin{aligned}\nu_{ijkl}^R(H) = {} & \nu_1^R[\epsilon_{imp}n_j n_k n_l + \epsilon_{jmp}n_i n_k n_l - \epsilon_{kmp}n_j n_i n_l \\ & -\epsilon_{lmp}n_j n_k n_i]\,n_p H_m \\ & + \nu_2^R[\epsilon_{jmp}n_l\delta_{ik} - \epsilon_{lmp}n_j\delta_{ik} + \epsilon_{jmp}n_k\delta_{il} \\ & -\epsilon_{kmp}n_j\delta_{il} + \epsilon_{imp}n_k\delta_{jl} - \epsilon_{kmp}n_i\delta_{jl} \\ & +\epsilon_{imp}n_l\delta_{jk} - \epsilon_{lmp}n_i\delta_{jk}]\,n_p H_m \\ & + \nu_3^R[\epsilon_{kmp}n_l\delta_{ij} + \epsilon_{lmp}n_k\delta_{ij} \\ & -\epsilon_{imp}n_j\delta_{kl} - \epsilon_{jmp}n_i\delta_{kl}]\,n_p H_m \\ & + \nu_4^R[\epsilon_{ikp}n_j n_l + \epsilon_{ilp}n_j n_k \\ & +\epsilon_{jlp}n_i n_k + \epsilon_{jkp}n_i n_l]\,n_p n_m H_m \\ & + \nu_5^R[\epsilon_{ikp}n_j n_l + \epsilon_{ilp}n_j n_k + \epsilon_{jlp}n_i n_k + \epsilon_{jkp}n_i n_l]\,H_p \\ & + \nu_6^R[\epsilon_{ikp}\delta_{jl} + \epsilon_{ilp}\delta_{jk} + \epsilon_{jlp}\delta_{ik} + \epsilon_{jkp}\delta_{il}]\,n_p n_m H_m \\ & + \nu_7^R[\epsilon_{ikp}\delta_{jl} + \epsilon_{ilp}\delta_{jk} + \epsilon_{jlp}\delta_{ik} + \epsilon_{jkp}\delta_{il}]\,H_p \\ & + \nu_8^R n_p[\epsilon_{ikp}(H_j n_l + H_l n_j) + \epsilon_{ilp}(H_j n_k + H_k n_j) \\ & +\epsilon_{jlp}(H_i n_k + H_k n_i) + \epsilon_{jkp}(H_i n_l + H_l n_i)]\end{aligned} \quad (25)$$

Because of $\nu_{ijkl}^R(H) = \nu_{jikl}^R(H)$ and $\nu_{ijkl}^R(H) = \nu_{ijlk}^R(H)$ the stress tensor remains symmetric and (25) does not contain any coupling to the vorticity. All contributions in (25) are reversible, where the antisymmetry w.r.t. exchange of the first pair of indices with the second one,

$\nu^R_{ijkl}(H) = \nu^R_{klij}(-H) = -\nu^R_{klij}(H)$, (according to Onsager's relation) guarantees zero entropy production. The major effect of these new terms is that a density wave (sound wave) is not only connected to div$\boldsymbol{v}$, but also to transverse velocities (and thus to all other variables). This will be manifest as (reversible) bulk shear stresses accompanying the sound wave. For example, for a density wave with amplitude $\Delta\rho$, frequency ω and wave vector k along the x-direction, magnetic field H along the y-direction, this bulk shear stress will be felt by a tracer particle as a transverse force

$$f_z = \bar{\nu}^R\,\omega\,k\,H\,\frac{\Delta\rho}{\rho_0} \tag{26}$$

with $\bar{\nu}^R = 2\nu^R_2{+}\nu^R_3{-}2\nu^R_7$, $\bar{\nu}^R = \nu^R_1{+}2\nu^R_2{-}\nu^R_3{-}2\nu^R_5{-}2\nu^R_7$, and $\bar{\nu}^R = -2\nu^R_6 - 2\nu^R_7$ if $\boldsymbol{n}$ is along the z-, x- and the y-direction, respectively.

3.5 Director reorientation

In eq.(7) the rotational viscosity $\gamma_1^{-1}\delta^\perp_{ij} \equiv (\gamma^{-1})_{ij}$ acquires linear field-dependent additions

$$(\gamma^{-1})^R_{ij}(H) = \frac{1}{\gamma^R_1}\epsilon_{ijk}n_k n_p H_p + \frac{1}{\gamma^R_2}(\epsilon_{ijp} + \epsilon_{ipk}n_k n_j - \epsilon_{jpk}n_k n_i)H_p \tag{27}$$

which are reversible and give zero entropy production, because they are antisymmetric in i and j according to Onsager's relation. Of course, they are also transverse to $\boldsymbol{n}$ in both indices $n_i\,(\gamma^{-1})^R_{ij}(H) = 0 = n_j\,(\gamma^{-1})^R_{ij}(H)$.

Usually, the so-called rotational viscosity γ_1 is measured by the homogeneous relaxation of the director towards an external magnetic field due to the magnetic anisotropy effect (13). In this case $\boldsymbol{H}$ and $\boldsymbol{n}(t)$ lie in the same plane all the time with relaxation rate $(\chi_a/\gamma_1)H^2$ [20]. The new terms in (27) change this picture. With (13,27) the director relaxation equation (3) takes the form

$$\dot{n}_i = \chi'_a\delta^\perp_{ij}H_j(\boldsymbol{H}\cdot\boldsymbol{n}) + \chi''_a(\boldsymbol{H}\times\boldsymbol{n})_i(\boldsymbol{H}\cdot\boldsymbol{n})^2 \tag{28}$$

where $\chi'_a = \chi_a/\gamma_1$ and $\chi''_a = \chi_a(1/\gamma^R_1{+}1/\gamma^R_2)$. Obviously, $\boldsymbol{n}(t)$ does not stay in the (initial) plane given by $\boldsymbol{H}$ and $\boldsymbol{n}(0)$, since there is a nonvanishing component $(\boldsymbol{H}\times\boldsymbol{n})\cdot\dot{\boldsymbol{n}}$ in (28). Thus, there are two distinct and coupled dynamic processes involved. For small angles the solutions of (28) can be written

$$\begin{aligned}\varphi &= \varphi_0\exp(-\chi'_a H^2 t)\cos(\chi''_a H^3 t)\\ \theta &= \varphi_0\exp(-\chi'_a H^2 t)\sin(\chi''_a H^3 t)\end{aligned} \tag{29}$$

where φ is the angle between the field and the projection of $\boldsymbol{n}(t)$ onto the initial plane $\boldsymbol{H}/\boldsymbol{n}(0)$ and θ is the angle of $\boldsymbol{n}(t)$ with this initial plane. The time dependence of φ is not a simple exponential decay, but shows an oscillation about it. The angle θ describes spatial oscillations of the director during the reorientation process. With field reversal $\boldsymbol{H} \to -\boldsymbol{H}$, also θ changes sign. Without the new reversible terms ($\chi''_a = 0$) a simple relaxation for φ, which is then the true angle between $\boldsymbol{n}$ and $\boldsymbol{H}$ is regained.

Appendix: Static relations

The statics of a macroscopic system is governed by its free energy. For nematics the free energy density in harmonic approximation reads [19–21]

$$f = \frac{T}{2C_V}(\delta\sigma)^2 + \frac{1}{2\rho^2\kappa_s}(\delta\rho)^2 + \frac{\gamma}{2}(\delta c)^2 + \frac{1}{\rho\alpha_s}(\delta\sigma)(\delta\rho) + \beta_\sigma(\delta c)(\delta\sigma) + \beta_\rho(\delta c)(\delta\rho) + \frac{1}{2}K_{ijkl}(\nabla_j n_i)(\nabla_l n_k) \tag{A.1}$$

where the Frank tensor

$$K_{ijkl} = K_1\delta^\perp_{ij}\delta^\perp_{kl} + K_2 n_p\epsilon_{pij}n_q\epsilon_{qkl} + K_3 n_j n_l\delta^\perp_{ik} \tag{A.2}$$

describes the energy cost for distorting the homogeneous alignment of the director. The conventional static susceptibilities contained in (A.1) are the specific heat (at constant density) C_V, the isentropic compressibility κ_s, the adiabatic volume expansion coefficient α_s and the appropriate susceptibilities γ, β_σ and β_ρ related to the concentration instead of the total mass density.

Due to the Gibbs relation (the local manifestation of the first and second law of thermodynamics)

$$df = \mu\,d\rho + T\,d\sigma + \mu_c\,dc + \Phi_{ij}\,d\nabla_j n_i + h'_i\,dn_i \tag{A.3}$$

the conjugate quantities follow from the free energy density by partial differentiation

$$\delta T \equiv \frac{\partial f}{\partial\sigma} = \frac{T}{C_V}\delta\sigma + \frac{1}{\rho\alpha_s}\delta\rho + \beta_\sigma\delta c \tag{A.4}$$

$$\delta\mu \equiv \frac{\partial f}{\partial\rho} = \frac{1}{\rho^2\kappa_s}\delta\rho + \frac{1}{\rho\alpha_s}\delta\sigma + \beta_\rho\delta c \tag{A.5}$$

$$\delta\mu_c \equiv \frac{\partial f}{\partial c} = \gamma\,\delta c + \beta_\sigma\delta\sigma + \beta_\rho\delta\rho \tag{A.6}$$

$$\Phi_{ij} \equiv \frac{\partial f}{\partial\nabla_j n_i} = K_{ijkl}\nabla_l n_k \tag{A.7}$$

$$h'_i \equiv \frac{\partial f}{\partial n_i} = \delta^\perp_{iq}\frac{\partial K_{pjkl}}{2\,\partial n_q}(\nabla_l n_k)(\nabla_j n_p) \tag{A.8}$$

The response to static director deformations is given by

$$h_i \equiv \frac{\delta}{\delta n_i}\int f dV = h'_i - \nabla_j\Phi_{ij} \tag{A.9}$$

The pressure is related to the other conjugate quantities by the Gibbs-Duhem relation [21]

$$\delta p = \rho\delta\mu + \sigma\delta T - \mu_c\delta c - h_j\delta n_j \tag{A.10}$$

neglecting contributions quadratic in the velocity.

The effect of an external magnetic field H on the statics is rather simple. Since all static equations have to be invariant under time reversal symmetry, and H changes sign under this symmetry, only quadratic contributions to the free energy are allowed in lowest order

$$f^{(M)} = -\frac{1}{2}\chi_a(\boldsymbol{H}\cdot\boldsymbol{n})^2 \tag{A.11}$$

which is the diamagnetic anisotropy energy [20] giving rise to the magnetic molecular field (13). Depending on the sign of χ_a the static orientation of $\boldsymbol{n}$ is either parallel or perpendicular to $\boldsymbol{H}$.

Conclusions and Perspectives

In this paper we have derived hydrodynamic equations for ferronematics in the limit that the magnetic degree of freedom has relaxed to its equilibrium value. When comparing the equations derived here with those of ordinary nematic liquid crystals, we find that there is no additional contribution to the static behaviour linear in the magnetic field. This situation changes completely, however, when one investigates dynamic coupling terms that are linear in the magnetic field between the various hydrodynamic variables.

The new dynamic effects predicted here come in four classes. In most nematics, for temperatures far above a smectic phase, one observes the phenomenon of flow alignment. A shear flow applied to a spatially homogeneous director field leads to a stationary configuration in which the director includes an angle with its original orientation - the flow alignment angle. In the case of usual nematics the director lies in the shear plane. Here we predict that an additional magnetic field *in* the shear plane applied to a ferronematic forces the director *out* of the shear plane due to the dynamic effects given here, which couple the director to the shear flow dissipatively.

Applying a temperature gradient to a nematic leads to a heat flux that has components parallel to the applied temperature gradient and parallel to the director. Here we suggest that an additional magnetic field orthogonal to the temperature gradient applied to a ferronematic leads to an additional reversible heat current that is perpendicular to both, the applied magnetic field and the temperature gradient.

In most hydrodynamic systems including ordinary nematics only viscous effects couple the various components of the velocity field. For a ferronematic there are, in addition, several terms coupling the three components of the velocity field reversibly. One of the consequences of these new contributions could be detected experimentally studying the effect of a sound wave propagating in $x-$direction, say, on a tracer particle also exposed to a magnetic field in $y-$direction. For this configuration we predict the occurrence of a force on this tracer particle in $z-$ direction, that is perpendicular to the plane spanned by the two applied fields.

Director reorientation is expected to change as well when switching from an ordinary nematic to a ferronematic. Here we have shown that the director reorientation picks up reversible contributions in addition to the usual director diffusion associated with the rotational viscosity γ_1. The new reversible contributions are predicted to lead to a relaxation oscillation when the director is reoriented in an external magnetic field in contrast to the simple relaxation observed for usual nematics.

Acknowledgements

Partial support of this work through the Schwerpunktsprogramm 1104 'Colloidal Magnetic Fluids' of the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

References

1. F. Brochard and P.G. de Gennes, *J. Phys. (France)* **31**, 691 (1970).
2. J. Rault, P.E. Cladis, J.P. Burger, *Phys. Lett.* **A 32**, 199 (1970).
3. C.F. Hayes, *Mol. Cryst. Liq. Cryst.* **36**, 245 (1976).
4. L. Liébert and A. Martinet, *J. Phys. (France) Lett.* **40**, 363 (1979).
5. S.-H. Chen and S.H. Chiang, *Mol. Cryst. Liq. Cryst.* **144**, 359 (1987).
6. S. H. Chen and N. M. Amer, *Phys. Rev. Lett.* **51**, 2298 (1983).
7. J.C. Bacri and A.M. Figueiredo-Neto, *Phys. Rev.* **E 50**, 3860 (1994).
8. S.I. Burylov and Y.L. Raikher, *Mol. Cryst. Liq. Cryst.* **258**, 123 (1995).
9. M. Koneracká, V. Závišová, P. Kopčanský, J. Jadżyn, G. Czechowski, B. Żywucki, *J. Magn. Magn. Mater.* **157/158**, 589 (1996).
10. S. Fontanini, A.L. Alexe-Ionescu, G. Barbero, A.M. Figueiredo Neto, *J. Chem. Phys.* **106**, 6187 (1997).
11. V. Berejnov, J.-C. Bacri, V. Cabuil, R. Perzynski and Y.L. Raikher, *Europhys. Lett.* **41**, 507 (1998).
12. A.Yu. Zubarev and L.Yu. Iskakova, *J. Magn. Magn. Mater.* **183**, 201 (1998).
13. Y.L. Raikher and V.I. Stepanov, *J. Magn. Magn. Mater.* **201**, 182 (1999).
14. I. Potočová, M. Koneracká, P. Kopčanský, M. Timko, L. Tomčo, J. Jadżyn, G.Czechowski, *J. Magn. Magn. Mater.* **196**, 578 (1999).
15. C.Y. Matuo and A.M. Figueiredo Neto, *Phys. Rev.* **E 60**, 1815 (1999).
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, *Landau description of ferrofluid to ferronematic phase transitions*, Magnetohydrodynamics, in print (2001).
18. T.C. Lubensky, *Mol. Cryst. Liq. Cryst.* **23**, 99 (1973).
19. P. C. Martin, O. Parodi and P.S. Pershan, *Phys. Rev.* **A6**, 2401 (1972).
20. P.G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon Press, Oxford, 1993).
21. 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, 1996) p. 15ff.
22. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions*, Benjamin, Reading Mass. (1975)
23. R.E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, New York, 1985).
24. L.D. Landau and E.M. Lifshitz, *Electrodynamics of Continuous Media*, (Pergamon, Oxford, 1984).