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Abstract. We present the derivation of the macroscopic equations for uniaxial ferronematic gels and elastomers. We deal with the superparamagnetic case, where no permanent magnetization is present and the anisotropy is provided by the nematic director. We include the magnetization as an independent dynamic degree of freedom. As a consequence special emphasis is laid on possible static and dynamic cross-couplings between magnetization and the non-magnetic degrees of freedom, as director reorientations, flow, elastic strains and relative rotations between director and the elastic network. In particular, we find reversible dynamic cross-couplings among rotations of the magnetization, the director, relative rotations, and deformational flow that allow for new possibilities to manipulate such materials. Application of simple (oscillatory) shear induces, in general, a finite magnetization normal to the shear plane and a relative rotation in the shear plane, whose amplitudes are linear in the shear rate. Induced magnetization, induced relative rotation and the director are mutually orthogonal, with the director aligned obliquely to the flow direction. This orientation is independent of the shear rate and is a material property.

1 Introduction

A major driving force behind the study of complex fluids and/or soft matter is the investigation of materials with novel properties. One already well-studied class of complex fluids are magnetic liquids [1], suspensions of magnetic monodomain particles whose properties can be tuned and controlled by fairly small magnetic fields [2–10]. The latter property led Brochard and de Gennes [11] early on to the suggestion to synthesize ferronematics; this generated numerous experimental activities in the field of liquid crystals including thermotropic, columnar and lyotropic ferronematic phases [12–22]. Macroscopic properties of ferronematics have been studied in [11–24]. Only last year, however, there was success in demonstrating ferromagnetic order in ferronematics [25].

In parallel Zrinyi's group synthesized and characterized isotropic magnetic gels, which showed large deformations under moderate external magnetic fields [26–34]. Later on physical properties in the macroscopic regime [35] as well as in the mesoscopic regime [36] have been examined. More recently this approach was generalized by generating uniaxial magnetic gels when the cross-linking was done in a magnetic field [37, 38] thus giving rise to a new class of anisotropic materials [37–39]. It possesses novel cross-coupling terms between the magnetic and the elas-

tomeric subsystems, which are frequently mediated by relative rotations between the elastic and magnetic degrees of freedom [40].

Another type of complex fluids with many applications in fields such as soaps and displays are liquid crystals. They are characterized by long-range orientational order, but for which positional order is only short range in 3D (nematics) or positional order occurs only in the form of density waves such as for smectic and columnar liquid crystals. In 1978 Finkelmann, Ringsdorf and Wendorff [41] combined the properties of low-molecular-weight liquid crystals with those of polymeric materials and synthesized side chain liquid crystalline polymers for which the mesogenic units (liquid crystalline building blocks) are attached via a flexible spacer to the polymeric backbones. This work led to the large field of liquid crystalline polymers. Stimulated by this development, Finkelmann's group started [42] and built up the field of liquid crystalline elastomers and gels [42–51] by cross-linking liquid crystalline polymers. This class of materials combines the properties of liquid crystals with those of rubbers and gels [50]. For example, applying a mechanical force leads to the reorientation of the director field and thus to a tunable change in optical properties [50]. It turns out that for these materials relative rotations between the network and the director field form a new category of macroscopic variables as suggested by de Gennes [52]. In the sequel the concept of relative rotations was incorporated into the macroscopic dynamic description of liquid crystalline elastomers [53]

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and it was shown that the nonlinear generalization of relative rotations [54] plays a crucial role in describing the nonlinear reorientation behavior of the nematic director under an external mechanical force [55–58].

The subject of the present paper is the investigation of the macroscopic behavior of ferronematic gels and elastomers. This type of materials possesses as additional macroscopic degrees of freedom compared to a simple fluid a magnetization density, the orientational degrees of freedom characteristic of a nematic, the strain field as it is associated with networks and gels, as well as relative rotations between the subsystems. Due to the long-range orientational order these systems are spatially anisotropic. Specifically we will study uniaxial ferronematic gels and elastomers as the simplest possibility. In addition, we will assume that the system does not order ferromagnetically (that is it does not have a permanent magnetization) and we will also restrict ourselves to the case of sufficiently weak external magnetic fields. The case of ferromagnetic order and/or strong external magnetic fields will be discussed elsewhere. Given the assumptions just outlined, we can draw on previous experience modeling the macroscopic behavior of nematic elastomers [53], ferronematics [23, 24], isotropic ferrogels [35] and uniaxial magnetic gels [40].

We find that there are particularly interesting reversible dynamic cross-coupling terms of the magnetization density to the molecular field associated with the director, and the thermodynamic conjugate to relative rotations. The case of an externally applied simple shear flow will be investigated in detail.

The present paper is organized as follows. In sect. 2 we describe the choice of the macroscopic variables, the statics and the thermodynamics. In sect. 3 we derive the resulting dynamic macroscopic equations. In sect. 4 we investigate some simple solutions of the macroscopic equations presented. Finally we present in sect. 5 a brief summary and conclusions. In appendix A we discuss selected static and dissipative higher-order gradient terms.

2 Statics and thermodynamics

To derive macroscopic equations for nematic ferrogels and elastomers we generalize suitably the macroscopic dynamics of ferronematics derived in ref. [24]. According to the Eulerian description all variables are local fields, *i.e.* volume densities that depend on space and time. As for ferronematics we have a number of variables associated with local conservation laws including density (ρ), density of linear momentum (\mathbf{g}), generalized energy density (ϵ) and the concentration of the magnetic particles (c). 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. The director variations $\delta\mathbf{n}$ of the director \mathbf{n} are associated with the spontaneously broken orientational symmetry. The director 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 . This variable is not associated with any conservation law, nor with a broken symmetry, but relaxes slowly to its equilibrium value. Due to the presence of a network the strain tensor ε_{ij} arises as a macroscopic variable; in its linearized version it takes the form $\varepsilon_{ij} = \frac{1}{2}(\nabla_i u_j + \nabla_j u_i)$ with the displacement field u_i . Due to the simultaneous presence of a network as well as of the variables δn_i , relative rotations $\tilde{\Omega}_i$ as pioneered by de Gennes [52] for nematic elastomers become an important macroscopic variable, which can be introduced via $\tilde{\Omega}_i = \delta n_i - \Omega_i^\perp$ as for the case of nematic liquid crystalline elastomers [53], where $\Omega_i^\perp = n_j \Omega_{ij}$ with $n_i \Omega_i^\perp = 0$, $\Omega_{ij} = \frac{1}{2}(\nabla_i u_j - \nabla_j u_i)$ and $n_i \delta n_i = 0$. Relative rotations are not truly hydrodynamic variables, but relax slowly enough to be considered here.

For the magnetic degrees of freedom we consider the case without a permanent magnetization \mathbf{M}_0 and no strong external field. The case of true ferromagnetism or a strong external field will be considered elsewhere. We thus have the magnetization M_i as a variable slowly relaxing to zero. To satisfy Maxwell's equations we take into account the magnetic induction \mathbf{B} . We use as a starting point the local formulation of the first law of thermodynamics [59–62] relating changes of the entropy density σ to all other macroscopic variables. The underlying assumption in this connection is that all microscopic variables are relaxing much faster and are in equilibrium already. We have

$$\begin{aligned} Td\sigma = & d\epsilon - \mu d\rho - \mu_c dc - v_i dg_i - W dS \\ & - H_i dB_i - h_i^M dM_i - h_i^{n'} dn_i - \Phi_{ij} d(\nabla_j n_i) \\ & - \psi_{ij} d\varepsilon_{ij} - L_i^\perp d\tilde{\Omega}_i, \end{aligned} \quad (1)$$

where all macroscopic variables are connected to the entropy density σ and where we have also taken into account the degree of nematic order, S in writing down eq. (1). The thermodynamic forces entering eq. (1) are 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 , the magnetic molecular field (h_i^M), the “order molecular field” (W), the elastic stress (ψ_{ij}) and the “relative molecular field” (L_i^\perp) associated with relative rotations.

The thermodynamic forces and thus simultaneously the static properties of nematic ferrogels and elastomers are obtained by expanding first the generalized energy density into the macroscopic variables and then, in a second step, by taking the variational derivatives with respect to one variable while keeping all other variables fixed [62]. We obtain

$$\epsilon = \epsilon_f + \epsilon_n + \epsilon_M + \epsilon_{nM}, \quad (2)$$

where ϵ_n contains the contributions of a usual nematic elastomer, ϵ_m is the magnetic energy density and ϵ_{nM} contains static cross-coupling terms between magnetic and elastomeric degrees of freedom. The energy density of a standard mixture of miscible fluids, ϵ_f [62], will not be

repeated here. For nematic elastomers, in the harmonic approximation

$$\begin{aligned} \epsilon_n = & \frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} + (\chi_{ij}^\rho\delta\rho + \chi_{ij}^\sigma\delta\sigma + \chi_{ij}^c\delta c + \chi_{ij}^S\delta S)\varepsilon_{ij} \\ & + \frac{1}{2}K_{ijkl}(\nabla_j n_i)(\nabla_l n_k) + \frac{1}{2}a(\delta S)^2 \\ & + M_{ijk}(\nabla_i n_j)(\nabla_k \delta S) + (b^\sigma\delta\sigma + b^\rho\delta\rho + b^c\delta c)\delta S \\ & + \frac{1}{2}L_{ij}(\nabla_i \delta S)(\nabla_j \delta S) + \frac{1}{2}D_1\tilde{\Omega}_i\tilde{\Omega}_i \\ & + \frac{1}{2}D_2(\delta_{ik}^\perp n_j + \delta_{ij}^\perp n_k)\tilde{\Omega}_i\varepsilon_{jk}, \end{aligned} \quad (3)$$

where the form of the material tensors is listed in [53].

For the magnetic energy density we have

$$\begin{aligned} \epsilon_M(\mathbf{B}, \mathbf{M}) = & \frac{1}{2}B^2 - \mathbf{M} \cdot \mathbf{B} \\ & + \frac{1}{2}A_1(\mathbf{n} \cdot \mathbf{M})^2 + \frac{1}{2}A_2M^2, \end{aligned} \quad (4)$$

where 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 experiments [18, 63, 64] 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. Only for a linearized description, valid for small deviations from equilibrium, are the coefficients constant.

For the coupling between the effects of the network and the magnetization we obtain (in close analogy to eq. (A2) of ref. [53])

$$\epsilon_{nM} = \frac{1}{2}\gamma_{ijkl}^M M_i M_j \varepsilon_{kl} + \gamma_{ijk}^{M\Omega} M_i M_j \tilde{\Omega}_k. \quad (5)$$

Here the term $\sim \gamma_{ijkl}^M$ corresponds to magnetostriction familiar from classical solids. In a uniaxial material one has six independent coefficients [65, 66]. The term $\sim \gamma_{ijk}^{M\Omega}$ is specific for systems with relative rotations. For uniaxial systems this cross-coupling takes the form

$$\gamma_{ijk}^{M\Omega} = \gamma^{M\Omega}(n_i\delta_{jk}^\perp + n_j\delta_{ik}^\perp) \quad (6)$$

and has thus only one independent coefficient.

The statics, *i.e.* the relation between the conjugate quantities and the variables, is now obtained by partial derivation. From eqs. (4) and (5), we find the magnetic Maxwell field H_i

$$H_i = \left(\frac{\partial \epsilon}{\partial B_i} \right)_{\mathbf{M}, \mathbf{n}, \dots} = B_i - M_i, \quad (7)$$

while the magnetic molecular field h_i^M reads

$$\begin{aligned} h_i^M = \left(\frac{\partial \epsilon}{\partial M_i} \right)_{\mathbf{B}, \mathbf{n}, \dots} = & -B_i + A_1 n_i n_j M_j + A_2 M_i \\ & + \gamma_{ijkl}^M M_j \varepsilon_{kl} + \gamma_{ijk}^{M\Omega} M_j \tilde{\Omega}_k. \end{aligned} \quad (8)$$

Note that because of the definition (7), 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 with the result $B_i = A_1 n_i n_j M_j + A_2 M_i + \gamma_{ijkl}^M M_j \varepsilon_{kl} + \gamma_{ijk}^{M\Omega} M_j \tilde{\Omega}_k$. With that solution the magnetic energy density (4) can be expressed as $\epsilon_M(\mathbf{B}) = \frac{1}{2}H_i(\mathbf{B})B_i$. 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 $\mu_{ij} = \mu_\perp \delta_{ij}^\perp + \mu_\parallel n_i n_j = \mu_\perp \delta_{ij} + \mu_a n_i n_j$ (where $\delta_{ij}^\perp = \delta_{ij} - n_i n_j$ is the transverse Kronecker symbol). Comparison with the equilibrium solution of (8) 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 (4) obtains the familiar form $\epsilon_M(\mathbf{B}) = \frac{1}{2}\mu_{ij}H_i(\mathbf{B})H_j(\mathbf{B})$. 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 $\epsilon_M(\mathbf{B})$ usually the Legendre-transformed energy $\epsilon_M(\mathbf{H}) = \epsilon_M(\mathbf{B}) - \mathbf{B} \cdot \mathbf{H}$ is used, for which we get the well-known form $2\epsilon_M(\mathbf{H}) = -\chi_a(\mathbf{n} \cdot \mathbf{H})^2 - (\chi_\perp - 1)\mathbf{H}^2$ where very often the vacuum field energy (\mathbf{H}^2) is omitted [67].

For the contribution to the molecular field $h_i^{n'}$ coming from the magnetic energy eq. (4) we have

$$h_i^{n'} = \delta_{ij}^\perp \left(\frac{\partial \epsilon}{\partial n_j} \right)_{\mathbf{M}, \mathbf{B}, \dots} = A_1 \delta_{ij}^\perp M_j M_k n_k \quad (9)$$

in addition to other (quadratic) nonlinear terms resulting from the orientation dependence of the material tensors in eqs. (3), *e.g.* $h_i^{n'} = D_2 \delta_{jk}^\perp \tilde{\Omega}_j \varepsilon_{ik}$ or [60] $h_i^{n'} = \frac{1}{2}(\partial K_{pqkl} / \partial n_i)(\nabla_j n_p)(\nabla_l n_k)$ etc. The Frank free energy density gives rise to

$$\Phi_{ij} = \left(\frac{\partial \epsilon}{\partial \nabla_j n_i} \right)_{\mathbf{M}, \mathbf{B}, \dots} = K_{ijkl} \nabla_k n_l, \quad (10)$$

which can be combined to the field

$$h_i^n \equiv h_i^{n'} - \nabla_j \Phi_{ij}. \quad (11)$$

Since \mathbf{n} is a unit vector that can only rotate, $n_i h_i^{n'} = 0$, which is ensured in (9) by the transverse Kronecker symbol. In equilibrium, $\mathbf{n} = \text{const.}$ and $\delta_{ij}^\perp M_j = 0$, hence $h_i^n = 0$.

Next we give the expressions for the thermodynamic forces in terms of the hydrodynamic and macroscopic variables (suppressing the standard fluid contributions)

$$\delta T = \left(\frac{\partial \epsilon}{\partial \delta \sigma} \right)_{\dots} = \chi_{ij}^\sigma \varepsilon_{ij} + b^\sigma \delta S, \quad (12)$$

$$\delta \mu = \left(\frac{\partial \epsilon}{\partial \delta \rho} \right)_{\dots} = \chi_{ij}^\rho \varepsilon_{ij} + b^\rho \delta S, \quad (13)$$

$$\delta \mu_c = \left(\frac{\partial \epsilon}{\partial \delta c_i} \right)_{\dots} = \chi_{ij}^c \varepsilon_{ij} + b^c \delta S, \quad (14)$$

$$\begin{aligned} \delta W = \left(\frac{\partial \epsilon}{\partial \delta S} \right) \dots &= \chi_{ij}^S \varepsilon_{ij} + b^\sigma \delta \sigma + b^\rho \delta \rho + b^c \delta c \\ &+ a \delta S - \nabla_i (L_{ij} \nabla_j \delta S) \\ &- \nabla_k (M_{ijk} \nabla_i n_j), \end{aligned} \quad (15)$$

$$\begin{aligned} \psi_{ij} = \left(\frac{\partial \epsilon}{\partial \varepsilon_{ij}} \right) \dots &= c_{ijkl} \varepsilon_{kl} + \chi_{ij}^\rho \delta \rho + \chi_{ij}^\sigma \delta \sigma + \chi_{ij}^S \delta S \\ &+ \chi_{ij}^c \delta c + \frac{1}{2} D_2 (\delta_{ik}^\perp n_j + \delta_{jk}^\perp n_i) \tilde{\Omega}_k \\ &+ \chi_{klij}^M M_k M_l, \end{aligned} \quad (16)$$

$$\begin{aligned} L_i^\perp = \left(\frac{\partial \epsilon}{\partial \tilde{\Omega}_i} \right) \dots &= D_1 \tilde{\Omega}_i + \frac{1}{2} D_2 (n_j \delta_{ik}^\perp + n_k \delta_{ij}^\perp) \varepsilon_{jk} \\ &+ \gamma_{kji}^{M\Omega} M_j M_k. \end{aligned} \quad (17)$$

3 Dynamics

3.1 Dynamic equations

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

$$\frac{\partial}{\partial t} \rho + \operatorname{div} \rho \mathbf{v} = 0, \quad (18)$$

$$\frac{\partial}{\partial t} \sigma + \operatorname{div} \sigma \mathbf{v} + \operatorname{div} \mathbf{j}^\sigma = \frac{R}{T}, \quad (19)$$

$$\frac{\partial}{\partial t} g_i + \nabla_j (v_j g_i + \delta_{ij} p - \psi_{ij} + \sigma_{ij}^{\text{th}} + \sigma_{ij}) = 0, \quad (20)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) n_i + (\mathbf{n} \times \boldsymbol{\omega})_i + Y_i = 0, \quad (21)$$

$$\rho \left(\frac{\partial}{\partial t} + v_j \nabla_j \right) c + \operatorname{div} \mathbf{j}^c = 0, \quad (22)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) S + Z = 0, \quad (23)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) \tilde{\Omega}_i + Y_i^\Omega = 0, \quad (24)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) \varepsilon_{ij} - A_{ij} + X_{ij}^\varepsilon = 0, \quad (25)$$

$$\left(\frac{\partial}{\partial t} + v_j \nabla_j \right) M_i + (\mathbf{M} \times \boldsymbol{\omega})_i + X_i = 0, \quad (26)$$

with $g_i = \rho v_i$ and [53, 68]

$$\begin{aligned} \sigma_{ij}^{\text{th}} &= -B_j H_i + \Phi_{kj} \nabla_i n_k + 2\psi_{jk} \varepsilon_{ki} \\ &- \frac{1}{2} (n_j h_i^n - n_i h_j^n) - \frac{1}{2} (M_j h_i^M - M_i h_j^M) \\ &- \frac{1}{2} (\tilde{\Omega}_j L_i^\perp - \tilde{\Omega}_i L_j^\perp), \end{aligned} \quad (27)$$

where $A_{ij} = \frac{1}{2} (\nabla_i v_j + \nabla_j v_i)$ is the deformational flow and $\omega_i = \frac{1}{2} \varepsilon_{ijk} \nabla_j v_k$ the vorticity. The thermodynamic pressure p is given by

$$p = -\epsilon + T\sigma + \mu\rho + \mathbf{g} \cdot \mathbf{v} + \mathbf{B} \cdot \mathbf{H}. \quad (28)$$

The parts of the currents shown explicitly in (18)-(27) are not material dependent, but are given by general symmetry and thermodynamic principles [62], like transformation behavior under translations (transport terms) or rotations (convective terms) and by the requirement of zero-entropy production ($R = 0$) of all those terms together with the isotropic pressure term in eq. (20).

Using the requirement [62]

$$\begin{aligned} \omega_{ij} (-M_i B_j + h_i^M M_j + h_i^n n_j + \Phi_{ki} \nabla_j n_k \\ + \nabla_k (n_j \Phi_{ik}) + L_i^\perp \tilde{\Omega}_j + 2\psi_{ki} \varepsilon_{kj}) = 0, \end{aligned} \quad (29)$$

for any constant antisymmetric matrix $\omega_{ij} = -\omega_{ji}$, which ensures the rotational invariance of the Gibbs relation, eq. (1), the non-symmetric part of the stress tensor, eq. (27), can be transformed as

$$\begin{aligned} 2\sigma_{ij}^{\text{th}} &= -(B_j H_i + B_i H_j) + \Phi_{ki} \nabla_j n_k + \Phi_{kj} \nabla_i n_k \\ &+ 2(\psi_{jk} \varepsilon_{ki} + \psi_{ik} \varepsilon_{kj}) + \nabla_k (n_j \Phi_{ik} - n_i \Phi_{jk}). \end{aligned} \quad (30)$$

Now σ_{ij}^{th} is either symmetric or a divergence of an antisymmetric part, which ensures angular momentum conservation. It can be brought into a manifestly symmetric form by some redefinitions [60].

The source term R/T in (19) is the entropy production, which has to be zero for reversible, and positive for irreversible processes. 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 , Y_i^Ω , Z , X_i , and X_{ij}^ε , associated with the temporal changes of the director, relative rotations, the nematic order, the magnetization, and the strain tensor, respectively, are given below. These phenomenological currents and quasi-currents can be split into reversible (superscript R) and dissipative parts (superscript D), where the former have the same time reversal behavior as the time derivative of the appropriate variable and must give $R = 0$, while the latter have the opposite behavior and give $R > 0$. The phenomenological part of the stress tensor σ_{ij} has to be symmetric guaranteeing angular momentum conservation. The dynamic equation for the energy density follows from eqs. (18)-(26) via eq. (1), and is not shown here.

We can use the static Maxwell equations

$$\operatorname{curl} \mathbf{H} = \operatorname{curl} (\mathbf{B} - \mathbf{M}) = 0, \quad \operatorname{div} \mathbf{B} = 0, \quad (31)$$

to determine \mathbf{B} , since we are not dealing with electromagnetic effects.

3.2 Reversible dynamics

If we make use of the symmetry arguments mentioned above and use the condition $R = 0$, we obtain the following

phenomenological expressions for the reversible currents up to linear order in the thermodynamic forces:

$$j_i^{\sigma R} = 0, \quad (32)$$

$$j_i^{cR} = 0, \quad (33)$$

$$X_{ij}^{\varepsilon R} = 0, \quad (34)$$

$$\sigma_{ij}^R = -\frac{1}{2}\lambda_{kji}h_k^n + \beta_{ij}W - \frac{1}{2}\lambda^\perp(L_i^\perp n_j + L_j^\perp n_i), \quad (35)$$

$$Z^R = \beta_{ij}A_{ij}, \quad (36)$$

$$Y_i^R = -\frac{1}{2}\lambda_{ijk}A_{jk} + \chi^R \epsilon_{ijk}n_j h_k^M, \quad (37)$$

$$Y_i^{\Omega R} = -\frac{1}{2}\lambda^\perp(\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j)A_{jk} + \tilde{\tau}^R \epsilon_{ijk}n_j h_k^M, \quad (38)$$

$$X_i^R = \chi^R \epsilon_{ijk}n_j h_k^n + \tilde{\tau}^R \epsilon_{ijk}n_j L_k^\perp. \quad (39)$$

The flow alignment tensor λ_{ijk} , well-known from ordinary nematics, has the usual form [60] $\lambda_{ijk} = \lambda(\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j)$. The coupling of relative director rotations to flow, provided by the λ^\perp terms, can already be found in nematic elastomers, as is the case for the coupling between compressional flow and (nematic) order denoted by $\beta_{ij} = \beta_\perp \delta_{ij}^\perp + \beta_\parallel n_i n_j$. The dynamic cross-coupling between magnetization and director reorientations, described by χ^R , is a hallmark of ferronematics, while the $\tilde{\tau}^R$ couplings are specific for ferronematic elastomers and gels. The physical meaning of these reversible couplings will be explored in sect. 4.

3.3 Irreversible dynamics and entropy production

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

$$\begin{aligned} R = & \frac{1}{2}\kappa_{ij}(\nabla_i T)(\nabla_j T) + \frac{1}{2}\nu_{ijkl}A_{ij}A_{kl} + \frac{1}{2}\kappa_W W^2 \\ & + \frac{1}{2}\gamma_{ij}(\nabla_k \psi_{ik})(\nabla_l \psi_{jl}) + D_{ij}^T(\nabla_j T)(\nabla_i \mu_c) \\ & + \lambda_{ij}^{WT}(\nabla_j W)(\nabla_i T) + \lambda_{ij}^{W\mu}(\nabla_j W)(\nabla_i \mu_c) \\ & + (\nabla_j \psi_{ij})(\zeta_{ik}^T \nabla_k T + \zeta_{ik}^c \nabla_k \mu_c + \zeta_{ik}^W \nabla_k W) \\ & + \frac{1}{2}D_{ij}(\nabla_i \mu_c)(\nabla_j \mu_c) + \frac{1}{2}b_{ij}^D h_i^M h_j^M + c_{ijk}^D h_i^M A_{jk} \\ & + \frac{1}{2\gamma_1} h_i^n \delta_{ij}^\perp h_j^n + \frac{1}{2}\zeta^\perp L_i^\perp L_j^\perp \delta_{ij}^\perp + \zeta_{12} L_i^\perp h_j^n \delta_{ij}^\perp. \quad (40) \end{aligned}$$

Here ν_{ijkl} is the uniaxial viscosity tensor [60] and κ_{ij} , D_{ij}^T 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 $b_{ij}^D = b_\perp^\perp \delta_{ij}^\perp + b_\parallel^\perp n_i n_j$. All the 2nd-rank tensors mentioned above have this form. The

third-rank tensor c_{ijk}^D is specific to ferronematics [24] 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. (40) is restricted by the positivity of the entropy production.

The dissipative parts of the currents then read

$$j_i^{\sigma D} = -\kappa_{ij}\nabla_j T - D_{ij}^T \nabla_j \mu_c - \lambda_{ij}^{WT} \nabla_j W - \zeta_{ij}^T \nabla_k \psi_{jk}, \quad (41)$$

$$j_i^{cD} = -D_{ij}\nabla_j \mu_c - D_{ij}^T \nabla_j T - \lambda_{ij}^{W\mu} \nabla_j W - \zeta_{ij}^c \nabla_k \psi_{jk}, \quad (42)$$

$$\sigma_{ij}^D = -\nu_{ijkl}^D A_{kl} - c_{kij}^D h_k^M, \quad (43)$$

$$Y_i^D = \frac{1}{\gamma_1} \delta_{ij}^\perp h_j^n - \zeta_{12} L_i^\perp, \quad (44)$$

$$Y_i^{\Omega D} = \zeta^\perp L_i^\perp - \zeta_{12} h_i^n, \quad (45)$$

$$X_i^D = b_{ij}^D h_j^M + c_{ijk}^D A_{jk}, \quad (46)$$

$$\begin{aligned} Z^D = & \kappa_W W - \nabla_j(\lambda_{ij}^{WT} \nabla_i T + \lambda_{ij}^{W\mu} \nabla_i \mu_c) \\ & - \nabla_k(\zeta_{ik}^W \nabla_j \psi_{ij}), \quad (47) \end{aligned}$$

$$\begin{aligned} X_{ij}^{\varepsilon D} = & -\frac{1}{2}[\nabla_j(\zeta_{ik}^T \nabla_k T + \zeta_{ik}^c \nabla_k \mu_c + \zeta_{ik}^W \nabla_k W \\ & + \gamma_{ik} \nabla_l \psi_{kl}) + (i \leftrightarrow j)]. \quad (48) \end{aligned}$$

4 Experimental considerations

4.1 Some consequences of reversible cross-coupling terms

In this section we discuss some consequences of reversible cross-coupling terms as they arise for ferronematic gels. These cross-couplings will typically show up in the response to external fields such as flow fields or an external magnetic field. We emphasize that they bring along reversible phenomenological transport coefficients (in contrast to the more familiar dissipative transport associated with eqs. (41)–(48)). They can assume a large range of values, since they are not restricted in magnitude or in sign. Thus, they are different in nature from the (also reversible) advective and convective terms and other symmetry-required contributions made explicit in eqs. (18)–(27). In the latter case the values of the coefficients (*e.g.* 1 or 1/2) are fixed and restricted in size. In addition, we stress that the reversible effects outlined here are strictly dynamic and not present statically.

A novel reversible dynamic cross-coupling term not familiar from isotropic ferrogels or nematic gels and elastomers is associated with the coefficient χ^R in eqs. (37) and (39). Focusing on the contributions due to χ^R they take the form

$$\frac{\partial}{\partial t} n_i = \dots - \chi^R \epsilon_{ijk} n_j h_k^M, \quad (49)$$

$$\frac{\partial}{\partial t} m_i = \dots - \chi^R \epsilon_{ijk} n_j h_k^n. \quad (50)$$

Two physical consequences are obvious from eqs. (49) and (50). An applied magnetic field leads (to linear order in the field strength) to a director rotation. And conversely, a torque on the director—for example, via an electric or a flow field—leads to a magnetization current. In the dispersion relation (for linear plane wave deviations from equilibrium) these terms add a propagating contribution to linear order $\omega \sim (\chi^R)^2 k$ in the wave vector.

A second reversible cross-coupling between relative rotations and the magnetization with the coefficient $\tilde{\tau}^R$ is specific for nematic gels and elastomers

$$\frac{\partial}{\partial t} \tilde{\Omega} = \dots - \tilde{\tau}^R \epsilon_{ijk} n_j h_k^M, \quad (51)$$

$$\frac{\partial}{\partial t} m_i = \dots - \tilde{\tau}^R \epsilon_{ijk} n_j L_k^\perp. \quad (52)$$

Equations (51) and (52) reveal two rather unusual consequences. First, temporal changes of the relative rotations are generated by an external magnetic field as an effect linear in the magnetic field strength. Second, the presence of relative rotations leads to a magnetization current. Since here only non-hydrodynamic variables are involved, these terms show up in the dispersion relation as a wave-vector-independent propagating contribution $\omega \sim (\tilde{\tau}^R)^2$.

The last effects to be discussed here are known from nematic elastomers. Specializing eqs. (35) and (38) to the terms of interest here, we obtain

$$\sigma_{ij}^R = \dots - \frac{1}{2} \lambda^\perp (L_i^\perp n_j + L_j^\perp n_i), \quad (53)$$

$$\frac{\partial}{\partial t} \tilde{\Omega} = \dots + \frac{1}{2} \lambda^\perp (\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j) A_{jk}. \quad (54)$$

The contributions associated with λ^\perp describe couplings between relative rotations and flow. More precisely, an extensional flow field (or any flow containing an extensional flow part such as simple shear) will lead to temporal changes of relative rotations. Only purely rotational flows will not affect the dynamics. On the other hand, statically generated relative rotations (*e.g.* by an external electric field) will lead to shear stresses via eq. (53).

4.2 The effect of simple shear flow

In ordinary nematic liquid crystals it is well known that an external simple shear can lead to a stationary reorientation of the director, which lies in the shear plane rotated by an angle ϕ with respect to the flow direction. This angle is independent of the flow rate and is given by the flow alignment parameter λ , cf. eqs. (35) and (37). It is a purely reversible effect, only the external shear flow produces viscous dissipation.

In the following we will show that a similar stationary flow alignment state can also exist for ferronematic elastomers and gels under simple shear. However, now not only is the director rotated in the shear plane, but in addition there is first a (stationary) relative rotation in the shear plane and perpendicular to the director and, second,

a finite magnetization perpendicular to the shear plane. The effect involves the flow alignment parameter and all three reversible transport parameters discussed in the previous section, but in addition also the appropriate dissipative transport parameters, which leads to an additional dissipation due to the internal variables involved.

To investigate the effect of simple shear we take the $x-y$ plane as the shear plane, with the shear $\Sigma = \frac{1}{2} \nabla_x v_y$ leading to $A_{xy} = \Sigma$ and $\omega_z = \Sigma$. We look for a stationary solution of the macroscopic dynamic equations. Therefore, all time derivatives vanish and the currents and quasi-currents have to vanish. The solution is assumed to be spatially homogeneous with the effect that the stress tensor is constant and $\frac{\partial}{\partial t} g_i = 0$, eq. (20), is automatically fulfilled. The scalar variables are not involved in this shear flow problem. The elastic degree of freedom will be disregarded for the moment. Thus we are left with the following three vector equations (for the director, the relative rotations and the magnetization, cf. eqs. (21), (26), (37)–(39) and (44)–(46))

$$0 = -\frac{1}{2} \lambda (\delta_{ix}^\perp n_y + \delta_{iy}^\perp n_x) \Sigma + \chi^R \epsilon_{ijk} n_j h_k^M + \frac{1}{\gamma_1} \delta_{ij}^\perp h_j^n - \zeta_{12} L_i^\perp + \frac{1}{2} \epsilon_{ijz} n_j \Sigma, \quad (55)$$

$$0 = -\frac{1}{2} \lambda^\perp (\delta_{ix}^\perp n_y + \delta_{iy}^\perp n_x) \Sigma + \tilde{\tau}^R \epsilon_{ijk} n_j h_k^M + \zeta^\perp L_i^\perp - \zeta_{12} h_i^n, \quad (56)$$

$$0 = \chi^R \epsilon_{ijk} n_j h_k^n + \tilde{\tau}^R \epsilon_{ijk} n_j L_k^\perp + b_{ij}^D h_j^M + c_{ixy}^D \Sigma + \epsilon_{ijz} M_j \Sigma. \quad (57)$$

Assuming the director to be tilted in the shear plane by a yet undetermined angle ϕ , there is *e.g.* $n_i = \delta_{ix} \cos \phi + \delta_{iy} \sin \phi$. The relative rotations have to be perpendicular to n_i and as an ansatz are assumed to be in the shear plane, hence $\tilde{\Omega}_i = \tilde{\Omega} (\delta_{ix} \sin \phi - \delta_{iy} \cos \phi)$. The magnetization is perpendicular to the shear plane (and perpendicular to n_i and $\tilde{\Omega}_i$) with $M_i = M \delta_{iz}$. The two amplitudes, M and $\tilde{\Omega}$ are further unknown quantities. From eqs. (8)–(11) and (17) one easily gets with these choices $h_i^n = 0$, $h_i^M = A_2 M \delta_{iz}$, and $L_i^\perp = D_1 \tilde{\Omega}_i$.

The set of eqs. (55)–(57) is now reduced to three linear algebraic equations

$$\chi^R A_2 M - \zeta_{12} D_1 \tilde{\Omega} = \frac{1}{2} (1 - \lambda \cos 2\phi) \Sigma, \quad (58)$$

$$\tilde{\tau}^R A_2 M + \zeta^\perp D_1 \tilde{\Omega} = -\frac{1}{2} \lambda^\perp \Sigma \cos 2\phi, \quad (59)$$

$$b_\perp^D A_2 M - \tilde{\tau}^R D_1 \tilde{\Omega} = -c_D \Sigma \cos 2\phi, \quad (60)$$

which produce the desired stationary and homogeneous flow alignment solution with

$$\cos 2\phi = \frac{\tilde{\tau}^R P_2}{2N}, \quad (61)$$

$$A_2 M = -\frac{\zeta^\perp P_4 + \frac{1}{2} \lambda^\perp P_2}{N} \Sigma, \quad (62)$$

$$D_1 \tilde{\Omega} = \frac{\tilde{\tau}^R P_4}{2N} \Sigma, \quad (63)$$

where $N = P_2P_3 - P_1P_4$ with

$$P_1 = \zeta_{12}\tilde{\tau}^R + \zeta^\perp\chi^R, \quad (64)$$

$$P_2 = -b_\perp^D\zeta^\perp - (\tilde{\tau}^R)^2, \quad (65)$$

$$P_3 = \frac{1}{2}\lambda\tilde{\tau}^R - \frac{1}{2}\lambda^\perp\chi^R, \quad (66)$$

$$P_4 = \frac{1}{2}b_\perp^D\lambda^\perp - c_D\tilde{\tau}^R. \quad (67)$$

The angle ϕ and therefore the orientations of the director, the relative rotations and the magnetization are independent of the shear rate Σ . The amplitudes of the magnetization and the relative rotations are directly proportional to Σ . All quantities depend on reversible as well as irreversible transport parameters. In particular, the amplitude $\tilde{\Omega}$ would vanish, if no dissipative transport parameters were present. As a consequence, the force to maintain the externally driven shear flow not only has to overcome the viscous dissipation $\frac{1}{2}\nu_\perp\Sigma^2$, but also contributions due to a finite h_i^M , and L_i^\perp according to eq. (40).

Up to here we have not considered the elastic degree of freedom. A constant external shear flow leads to strain deformation, eq. (25), $\varepsilon_{xy} = \Sigma t$ that grows indefinitely with time. Therefore, constant shear flow can only be applied for a short time and is problematic experimentally. On the other hand, an oscillating shear with a periodically changing shear amplitude, $A_{xy} = \Sigma \cos \omega_S t$, does not influence the orientation of the director (and the magnetization and the relative rotations). If the shear frequency ω_S is small enough, such that the adjustment of the amplitudes M and $\tilde{\Omega}$ are faster than the changes in the shear amplitude, the amplitudes follow adiabatically the time dependence of the shear amplitude and the solution eqs. (62) and (63) are still valid with Σ replaced by $\Sigma \cos(\omega_S t)$.

There is, however, a further complication, when the shear is oscillatory. There is an energetic coupling ($\sim D_2$) between elastic strains and relative rotations, eq. (17), that makes the connection between $\tilde{\Omega}_i$ and L_i^\perp more complicated. Since the additional contribution $\sim D_2\Sigma(\sin \omega_S t/\omega_S)$ is out-of-phase with the external shear flow (and has an angle dependence different from that of L_i^\perp), the oscillation of the amplitude $\tilde{\Omega}$ shows a phase shift relative to the external shear flow (and is angle dependent). The relative rotation amplitude takes the form $\tilde{\Omega} \approx (\alpha/D_1)\Sigma \cos \omega_S t + (D_2/2D_1) \cos 2\phi \Sigma(\sin \omega_S t/\omega_S)$, where $\alpha = \tilde{\tau}^R P_4/2N$ corresponds to the static solution, eq. (63). The additional term gives the phase shift that is angle dependent. Of course, the tilt angle ϕ is time and shear rate independent and still given by eq. (61).

5 Summary and conclusions

In this paper we have presented the macroscopic dynamic equations for ferronematic gels and elastomers. We find that in comparison to nematic gels and to isotropic ferrogels a number of new cross-coupling terms arise, which might lead to applications. These are due, in particular, to

the coupling between the magnetization on the one hand and the director field and the relative rotations between the network and the director on the other. As an example we show how an applied oscillatory shear flow can give rise to a stationary nematic alignment and simultaneously to relative rotations within the shear plane and a magnetization perpendicular to it.

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Appendix A. Higher-order gradient terms

Quite recently there has been an increasing interest in higher-order gradient terms in the context of the dynamic density functional theory for colloidal liquid crystal phases [69–72]. Stimulated by these new developments in the field of colloidal physics, we discuss in the following selected higher-order gradient terms in statics and dissipative dynamics for the system studied in the present paper.

For the generalized energy we have

$$\begin{aligned} \epsilon_{hgt} = & N_{ijk}^\rho(\nabla_i\rho)(\nabla_j n_k) + N_{ijk}^\sigma(\nabla_i\sigma)(\nabla_j n_k) \\ & + N_{ijk}^c(\nabla_i c)(\nabla_j n_k) + N_{ijk}^S(\nabla_i\delta S)(\nabla_j n_k) \\ & + N_{ijk}^{\rho\Omega}(\nabla_i\rho)(\nabla_j\tilde{\Omega}_k) + N_{ijk}^{\sigma\Omega}(\nabla_i\sigma)(\nabla_j\tilde{\Omega}_k) \\ & + N_{ijk}^{c\Omega}(\nabla_i c)(\nabla_j\tilde{\Omega}_k) + N_{ijk}^{S\Omega}(\nabla_i\delta S)(\nabla_j\tilde{\Omega}_k), \end{aligned} \quad (A.1)$$

where all third-rank tensors take the form

$$N_{ijk}^\xi = N^\xi(\delta_{ik}^\perp n_j + \delta_{jk}^\perp n_i). \quad (A.2)$$

Higher-order gradient terms of the type of the first four terms in eq. (A.1) have been discussed first for low-molecular-weight nematic liquid crystals [73, 74]. The four last terms represent a new class of higher-order gradient terms given here for the first time. These terms are coupling gradients of the density, the entropy density, the concentration and of the order parameter variation to gradients of the relative rotations, $\tilde{\Omega}_i$. This new class of higher-order gradient terms not only exists for nematic ferrogels, but also for nematic liquid crystalline gels and elastomers.

It turns out that relative rotations also couple to gradients of several macroscopic variables dissipatively. Dissipative higher-order gradient terms supplementing eq. (40) take the form

$$\begin{aligned} R_{hgt} = & \zeta_{ijk}^{T\Omega}(\nabla_i T)(\nabla_j L_k^\perp) + \zeta_{ijk}^{\mu_c\Omega}(\nabla_i \mu_c)(\nabla_j L_k^\perp) \\ & + \zeta_{ijk}^{W\Omega}(\nabla_i W)(\nabla_j L_k^\perp), \end{aligned} \quad (A.3)$$

where the third-rank tensors $\zeta_{ijk}^{\xi\Omega}$ take the form

$$\zeta_{ijk}^{\xi\Omega} = \zeta^{\xi\Omega}(\delta_{ik}^\perp n_j + \delta_{jk}^\perp n_i). \quad (A.4)$$

Dissipative currents derived using these higher-order gradient terms enter the resulting macroscopic dynamic

equations at the same order in the gradients as the static higher-order gradient terms discussed above.

As an example of a reversible higher-order gradient contribution we present a coupling specific for magnetic gels representing a coupling between spatial variations of the magnetization and of the strain

$$X_{ij}^{\varepsilon R} = \frac{1}{2} \lambda_{lp}^M (\epsilon_{jkp} \nabla_i + \epsilon_{ikp} \nabla_j) \nabla_k h_l^M, \quad (\text{A.5})$$

$$X_i^R = \lambda_{ip}^M \epsilon_{pj k} \nabla_j \nabla_l \psi_{kl}, \quad (\text{A.6})$$

with $\lambda_{ij}^M = \lambda_{\parallel}^M n_i n_j + \lambda_{\perp}^M \delta_{ij}^{\perp}$. This coupling between inhomogeneous magnetization and inhomogeneous strains exists in isotropic magnetic gels as well (with $\lambda_{\perp}^M = \lambda_{\parallel}^M$). The λ^M terms yield reversible propagating contributions to the dispersion relations $\omega^2 \sim k^4$ and are therefore most likely masked by dissipation.

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