Macroscopic behavior of ferrocholesteric liquid crystals and ferrocholesteric gels and elastomers

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Abstract. We study the influence of macroscopic chirality on the macroscopic properties of superparamagnetic liquid crystals and gels. Specifically we derive macroscopic dynamic equations for ferrocholesteric low molecular weight (LMW) liquid crystals and for ferrocholesteric gels and elastomers in the local description using the director field as macroscopic variable. The magnetization is treated as a macroscopic dynamic degree of freedom and its coupling to all other macroscopic variables is examined in detail. We incorporate into our dynamic analysis terms that are linear in a magnetic field giving rise to a number of cross-coupling terms not possible otherwise. A number of properties that are unique to the class of systems studied arise. As an example for a static property we find a term in the generalized energy which is linear in the electric field and quadratic in the magnetic field. We find that applying a magnetic field to a ferrocholesteric can lead to reversible electric currents, heat currents and concentration currents, which change their sign with a sign change of macroscopic chirality. As an example of a rather intriguing dissipative dynamic contribution we point out that for ferrocholesterics and for ferrocholesteric gels and elastomers in a magnetic field extensional flow leads to electric and heat currents.

1 Introduction

The first liquid crystalline phase discovered were cholesteric liquid crystals [1]. Early on it was recognized [2] that this phase has rather unique dynamic properties such as a rotation of its superstructure in droplets in a temperature gradient [2]. Cholesteric liquid crystals are characterized by a macroscopic helical superstructure breaking mirror and thus parity symmetry macroscopically [3,4]. As a consequence low molecular weight (LMW) cholesteric liquid crystals possess a large number of rather unique optical and flow properties (compare refs. [3] and [4] for a review).

Over the decades, however, other liquid crystalline phases formed by LMW materials such as nematics (which have 3 D long range orientational order), smectics and columnar phases (showing density waves in one and two spatial dimensions, respectively) attracted more attention by chemists and physicists, in particular due to their large scale applications in fields like soaps (lyotropic multi-component systems) and liquid crystals displays (LCDs).

The properties of LMW liquid crystal phases were combined with those of another large class of complex fluids, namely polymers, when in 1978 Finkelmann, Ringsdorf and Wendorff [5] 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 and started the field of liquid crystalline sidechain polymers. Shortly thereafter Finkelmann and his collaborators started [6] and generated the field of liquid crystalline elastomers and gels [6–13] by cross-linking liquid crystalline polymers. Liquid crystalline elastomers and gels combine the properties of liquid crystals with those of rubbers and gels [12]. De Gennes pointed out that for this class of materials relative rotations between the network and the director field form a new category of macroscopic variables [14]. Over the last two decades or so relative rotations as macroscopic variables were incorporated into the macroscopic dynamic description of liquid crystalline elastomers [15]. In addition, Menzel et al. demonstrated that the nonlinear generalization of relative rotations [16] plays a crucial role in describing the nonlinear reorientation behavior of the nematic director under an external mechanical force [17–20].

Magnetic liquids [21], suspensions of magnetic monodomain particles whose properties can be tuned and controlled by fairly small magnetic fields [22–30], constitute another class of complex fluids, which has been investigated in quite some detail already. Because of its large response to small magnetic fields, Brochard and de Gennes [31] suggested to synthesize ferronematics combining the properties on nematics and magnetic liquids. In the following there was a number of experiments combining these magnetic liquids and various types of liquid

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crystalline phases [32–40]. These experiments also led to the investigation of the macroscopic properties of the combination of nematic LMWs and magnetic liquids [31–42]. Essentially all experimental studies in this field gave rise to materials which were superparamagnetic just as the usual magnetic liquids [21]. Quite recently, however, Mertelj et al. were able to provide convincing evidence experimentally of ferromagnetic order in a new type of ferronematics [43, 44].

Quite similarly as for liquid crystals and liquid crystalline elastomers, Zrinyi's group combined magnetic liquids and gels to synthesize and characterize isotropic magnetic gels, which reveal substantial deformations under moderate external magnetic fields [45–48]. Physical properties in the macroscopic regime [49] as well as in the mesoscopic regime [50] have also been characterized. Later uniaxial magnetic gels were produced when the cross-linking was done in a magnetic field [51, 52] thus giving rise to a new class of anisotropic materials [51–53]. For these uniaxial materials relative rotations between the elastic and magnetic degrees of freedom turn out to be of crucial importance [54, 55].

In the present paper we investigate the macroscopic behavior of ferrocholesterics as well as of ferrocholesteric gels and elastomers making use of the local description in which the director is used as a macroscopic variable. In contrast to the strictly hydrodynamic description [56], for which there is only one truly hydrodynamic variable, the director used in the local description in the literature [3, 4] also contains one macroscopic quantity, which relaxes on a long, but finite time scale [56–58]. Therefore we will use the same set of macroscopic variables as for the ferronematic case discussed recently [59], 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. Based on these assumptions, we can make use of our previous work modeling the macroscopic behavior of nematic elastomers [15], ferronematics [41, 42], isotropic ferrogels [49], uniaxial magnetic gels [54] and ferronematic gels and elastomers [59].

We point out that the equations derived contain as a special case LMW ferrocholesterics as well as cholesteric gels and elastomers. For these two classes of materials a complete set of macroscopic dynamic equations has not been given before either. While there appear to be no experimental studies as yet on ferrocholesteric gels and elastomers, ferrocholesteric LMWs [60–67] (and references therein) and cholesteric elastomers [12, 68–72] (and references therein) have been the subject of experimental and theoretical investigations already.

We find here that there is a number of novel cross-coupling terms in all three domains: statically as well as for the reversible and the irreversible currents. They are all intrinsically connected to the existence of the pseudoscalar quantity q_0 associated with the helical superstructure. In some cases we also find that terms which are in addition linear in a magnetic field allow for intriguing coupling terms, for example between the molecular field of the director and electric and heat currents.

The present paper is organized as follows. In section 2 we describe the choice of the macroscopic variables, the statics and the thermodynamics. In section 3 we derive the resulting dynamic macroscopic equations. In section 4 we investigate some simple solutions of the macroscopic equations presented. Finally we present in section 5 a brief summary and conclusions. In two appendices we give reversible and dissipative dynamic contributions to the macroscopic dynamics of ferrocholesterics and of ferronematics, which are linear in a magnetic field. This type of terms has never been discussed before for gels and elastomers. In a third appendix we discuss how the terms related to broken mirror symmetry influence the rheologically relevant macroscopic quantities elastic stress and stress tensor.

2 Statics and thermodynamics

To derive macroscopic equations for ferrocholesterics and ferrocholesteric gels and elastomers we generalize suitably the macroscopic dynamics of ferronematics derived in ref. [42] and for ferronematic gels and elastomers given recently [59]. Here we concentrate on the chiral aspects that are manifest by the existence of a pseudoscalar quantity, q_0, in the cholesteric phases. Under spatial inversion q_0 changes sign meaning that after inversion the phase is not identical to the original one. This handedness is the hallmark for chirality. The parameter q_0 is considered a given material property of the cholesteric phases, and it can depend on the (scalar) state variables, like temperature, density or concentration. It is not necessarily related to a molecular chirality ref. [72], but can also be introduced by a low symmetry structure in a given phase that allows both types of handedness to exist and to be energetically equivalent, a feature called ambidextrous chirality [73, 74].

According to the Eulerian description we use local fields, i.e. volume densities that depend on space and time, as dynamic state variables. Some of these variables are associated with local conservation laws including for ferrocholesterics mass density (ρ), density of linear momentum ($\boldsymbol{g}$), 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.

Here we use the local description of cholesteric liquid crystals making use of the director field $\boldsymbol{n}(\boldsymbol{r}, t)$ [3, 56, 57]. While the director variations $\delta\boldsymbol{n}$ are associated with the two spontaneously broken orientational symmetries in a nematic, only one of them is strictly hydrodynamic in a cholesteric liquid crystal [56]. Nevertheless both independent components of the director are macroscopic variables as long as the pitch of the cholesteric helix is sufficiently large compared to the molecular length. The director is a unit vector and due to its special symmetry all equations have to be invariant under the replacement $\boldsymbol{n} \rightarrow -\boldsymbol{n}$.

The magnetization $\boldsymbol{M} = M\boldsymbol{m}$ is associated with rotations $\delta\boldsymbol{m}$ 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 [14] for nematic elastomers become an important macroscopic variable, which can also be introduced via $\tilde{\Omega}_i = \delta n_i - \Omega_i^\perp$ as for the case of nematic liquid crystalline elastomers [15], where $\Omega_i^\perp = n_j \Omega_{ij}$ with $n_i \Omega_i^\perp = 0$, $\Omega_{ij} = \frac{1}{2}(\nabla_i u_j - \nabla_i u_j)$ 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 $\boldsymbol{M}_0$. We also assume external fields, electric or magnetic, to be weak, in the sense that they do not change the symmetry of the phase. The case of true ferromagnetism or a strong magnetic 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 $\boldsymbol{B}$. To accommodate external electric fields we use the electric displacement field $\boldsymbol{D}$.

We use as a starting point the local formulation of the first law of thermodynamics [58,75–77] 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

$$
\begin{aligned}
T d\sigma = \ & d\varepsilon - \mu d\rho - \mu_c dc - v_i dg_i - W dS \\
& - H_i dB_i - h_i^M dM_i - h_i^{n\prime} dn_i - \Phi_{ij} d(\nabla_j n_i) \\
& - \psi_{ij} d\varepsilon_{ij} - L_i^\perp d\tilde{\Omega}_i - E_i dD_i
\end{aligned}
\tag{1}
$$

where all macroscopic variables are connected to the entropy density σ and where we have also taken into account the degree of orientational 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}, $h_i^{n\prime}$), 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, and the electric field (E_i).

The thermodynamic forces and thus simultaneously the static properties of ferrocholesterics and ferrocholesteric gels 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 [58].

We will not repeat the derivation of the static properties of (achiral) ferronematic elastomers, which can be found in Ref. [59]. Only those parts that are needed for the discussion in Sec. 4 will be shown there. We only want to mention that there are (achiral) static crosscouplings

between the strain and all scalar variables (including the nematic order) as well as the relative rotations, already on the harmonic energy level. In cubic order the magnetization is coupled statically to the director orientation, the relative rotations and the strains.

Here we discuss the chiral part of the energy, which comes about due to the existence of a pseudoscalar quantity, q_0, in cholesterics. In any system with a nematic order chirality allows for a linear twist energy that leads to a helical structure of the director [3]. In addition, the twist couples to all scalar variables (static Lehmann effects) [72,78,79] and to strains and relative rotations

$$
\begin{aligned}
\varepsilon_{ch} = \ & q_0 L_2\, \boldsymbol{n}\cdot(\boldsymbol{\nabla} \times \boldsymbol{n}) \\
& - q_0(\tau_c \delta c + \tau_\sigma \delta\sigma + \tau_\rho \delta\rho + \tau_S \delta S)\, \boldsymbol{n}\cdot(\boldsymbol{\nabla} \times \boldsymbol{n}) \\
& - q_0 \tau_{ij}^\varepsilon\, \boldsymbol{n}\cdot(\boldsymbol{\nabla} \times \boldsymbol{n})\, \delta\varepsilon_{ij} - q_0 \tau_\Omega \epsilon_{ikp} n_j n_p \tilde{\Omega}_i \nabla_j n_k
\end{aligned}
\tag{2}
$$

The pitch of the helix generally is $\sim q_0$ and becomes exactly q_0, if the coefficient of the linear twist term, L_2, is identical to the quadratic twist elastic coefficient K_2 [80]. The helical axis $\boldsymbol{p}$ is perpendicular to the rotation plane of the director in the cholesteric case. The contribution $\sim \tau_S$ is associated with variations of the modulus of the orientational order parameter and arises for all cholesteric systems.

The contributions in the last line of eq.(2) are specific for cholesteric elastic systems and have not been given before. The first one represents a coupling of twist to the strain tensor ε, where τ_{ij}^ε takes the form

$$
\tau_{ij}^\varepsilon = \tau_1^\varepsilon n_i n_j + \tau_2^\varepsilon \delta_{ij}^\perp
\tag{3}
$$

Thus this term gives rise to changes in the pitch due to uniaxial mechanical stresses such as compression and dilatation. This effect has been studied for cholesteric sidechain elastomers in detail experimentally [70,71]. The second one ($\sim \tau_\Omega$) relates static director deformations with relative rotations.

Other static effects specific for general cholesteric elastic systems are related to electric fields

$$
\varepsilon_{Dch} = q_0 \zeta^\Omega n_j \epsilon_{ijk} D_i \tilde{\Omega}_k + q_0 \zeta_{ijk}^\psi D_i \varepsilon_{jk}
\tag{4}
$$

and describe electric field-induced relative rotations (rotato-electricity [68,69]) and deformations with

$$
\zeta_{ijk}^\psi = \zeta^\psi(\epsilon_{ijp} n_p n_k + \epsilon_{ikp} n_p n_j)
\tag{5}
$$

Since the magnetization and/or the magnetic field is odd under time reversal, there are no static linear magnetic field effects.

For magnetic cholesteric elastic systems there are additional static couplings, if a magnetic field is present

$$
\varepsilon_{Mch} = q_0 H_i (F_{ijk} M_j D_k + G_{ijkl} M_j \nabla_k n_l)
\tag{6}
$$

coupling the magnetization to electric fields and director deformations. The material tensor F_{ijk} is of the form

$$
\begin{aligned}
F_{ijk}^{\alpha\beta} = \ & F_1 \epsilon_{ijk} + F_2 \epsilon_{ijp} n_p n_k \\
& + F_3 \epsilon_{ipk} n_p n_j + F_4 \epsilon_{pjk} n_p n_i
\end{aligned}
\tag{7}
$$

while for G_{ijkl} one gets

$$
\begin{aligned}
G_{ijkl} = {}& n_p\epsilon_{lpi}(G_1 n_j n_k + G_2\delta_{jk}^\perp) \\
& + n_p\epsilon_{lpj}(G_3 n_i n_k + G_4\delta_{ik}^\perp) \\
& + n_p\epsilon_{lpk}(G_5 n_i n_j + G_6\delta_{ij}^\perp) \\
& + \delta_{lp}^\perp(G_7 n_k\epsilon_{ijp} + G_8 n_j\epsilon_{ikp} + G_9 n_i\epsilon_{jkp}) \quad (8)
\end{aligned}
$$

Next we give the additional chiral contributions to the thermodynamic conjugate quantities that arise from the chiral energy $\varepsilon = \varepsilon_{ch} + \varepsilon_{Dch} + \varepsilon_{Mch}$, eqs.(2) -(6), by taking the variational derivative with respect to the appropriate variables

$$\delta T = -q_0\tau_\sigma\boldsymbol{n}\cdot(\boldsymbol{\nabla}\times\boldsymbol{n}) \tag{9}$$

$$\delta\mu = -q_0\tau_\rho\boldsymbol{n}\cdot(\boldsymbol{\nabla}\times\boldsymbol{n}) \tag{10}$$

$$\delta\mu_c = -q_0\tau_c\boldsymbol{n}\cdot(\boldsymbol{\nabla}\times\boldsymbol{n}) \tag{11}$$

$$W = -q_0\tau_S\boldsymbol{n}\cdot(\boldsymbol{\nabla}\times\boldsymbol{n}) \tag{12}$$

$$\psi_{ij} = -q_0\tau_{ij}^\varepsilon\boldsymbol{n}\cdot(\boldsymbol{\nabla}\times\boldsymbol{n}) + q_0\zeta_{kij}^\psi D_k \tag{13}$$

$$L_i^\perp = +q_0\epsilon_{jki}(\zeta^\Omega D_j + \tau_\Omega n_j n_p\nabla_p)n_k \tag{14}$$

$$E_i = q_0\zeta^\Omega n_j\epsilon_{ijk}\tilde{\Omega}_k + q_0 H_k F_{kji}M_j + q_0\zeta_{ijk}^\psi\varepsilon_{jk} \tag{15}$$

$$h_i^M = q_0 H_j F_{jik}D_k \tag{16}$$

$$
\begin{aligned}
h_i^n = {}& q_0 L_2(\boldsymbol{\nabla}\times\boldsymbol{n})_j\delta_{ij}^\perp + q_0\tau_\Omega n_j n_p\epsilon_{kip}\nabla_j\tilde{\Omega}_k \\
& + q_0 n_k\epsilon_{kji}(\tau_c\nabla_j c + \tau_\sigma\nabla_j\sigma + \tau_\rho\nabla_j\rho + \tau_S\nabla_j S) \\
& + q_0\tau_{kl}^\varepsilon n_p\epsilon_{pji}\nabla_j\varepsilon_{kl} - q_0 H_k G_{klji}\nabla_j M_l \quad (17)
\end{aligned}
$$

In section 4 we will suggest experiments to observe the consequences of some of these new static chiral terms.

3 Dynamics

3.1 Dynamic equations

The dynamic macroscopic variables for ferronematic gels and elastomers on the one hand and for cholesteric gels and elastomers in their local description on the other hand are the same. Consequently the balance equations for conserved quantities, broken symmetry variables and slowly relaxing variables have the same overall structure [59]. Therefore the discussion in the present subsection closely parallels that of section 3.1 in ref. [59]. For low molecular weight ferrocholesterics the dynamic equations for the stress tensor, ε_{ij}, and for the relative rotations, $\tilde{\Omega}_i$, are absent. In addition there is also no dynamic equation for the magnetization in the case of usual low molecular weight cholesteric liquid crystals. Therefore we have

$$\frac{\partial}{\partial t}\rho + \operatorname{div}\rho\boldsymbol{v} = 0 \tag{18}$$

$$\frac{\partial}{\partial t}\sigma + \operatorname{div}\sigma\boldsymbol{v} + \operatorname{div}\boldsymbol{j}^{\boldsymbol{\sigma}} = \frac{R}{T} \tag{19}$$

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

$$\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)n_i + (\boldsymbol{n}\times\boldsymbol{\omega})_i + Y_i = 0 \tag{21}$$

$$\rho\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)c + \operatorname{div}\boldsymbol{j}^c = 0 \tag{22}$$

$$\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)S + Z = 0 \tag{23}$$

$$\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)\tilde{\Omega}_i + Y_i^\Omega = 0 \tag{24}$$

$$\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)\varepsilon_{ij} - A_{ij} + X_{ij}^\varepsilon = 0 \tag{25}$$

$$\Big(\frac{\partial}{\partial t} + v_j\nabla_j\Big)M_i + (\boldsymbol{M}\times\boldsymbol{\omega})_i + X_i = 0 \tag{26}$$

with $g_i = \rho v_i$. Using the requirement of the rotational invariance of the energy one can write [15, 58, 81]

$$
\begin{aligned}
2\sigma_{ij}^{th} = {}& -(D_j E_i + D_i E_j) - (B_j H_i + B_i H_j) \\
& + \Phi_{ki}\nabla_j n_k + \Phi_{kj}\nabla_i n_k + \nabla_k(n_j\Phi_{ik} - n_i\Phi_{jk}) \\
& + 2(\psi_{jk}\varepsilon_{ki} + \psi_{ik}\varepsilon_{kj}) \quad (27)
\end{aligned}
$$

where 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 [76].

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, have been presented in detail for ferronematic gels and elastomers in ref. [59]. We will introduce them only, when needed below.

The additions brought about by the existence of the pseudoscalar quantity q_0 to the phenomenological currents and quasi-currents can (also) 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.

For the reversible and irreversible currents we will focus on the lowest order in the gradients. In addition, we will present reversible and dissipative dynamic contributions, which are linear in an external magnetic field, $\boldsymbol{H}$, in a separate appendix.

We can use the static Maxwell equations

$$\mathrm{curl}\,\boldsymbol{H} = \mathrm{curl}\,(\boldsymbol{B} - \boldsymbol{M}) = 0, \qquad \mathrm{div}\,\boldsymbol{B} = 0, \qquad (28)$$

to determine $\boldsymbol{B}$, since we are not dealing with electromagnetic effects. For the electrostatic Maxwell fields we take $\mathrm{curl}\,\boldsymbol{E} = 0$ and $\mathrm{div}\,\boldsymbol{D} = \rho_e$. Conservation of the electric charge ρ_E is expressed by $(\partial/\partial t)D_i + v_k\nabla_k D_i + (\boldsymbol{D} \times \boldsymbol{\omega})_i + j_i^e = 0$ with j_i^e the electric current density.

3.2 Reversible dynamics

Using the condition $R = 0$ and the general symmetry arguments outlined above, we obtain the following expressions for the chiral parts of the reversible currents up to linear order in the thermodynamic forces without a magnetic field H_i

$$j_i^{\sigma R} = q_0(\chi_{ijk}^{\sigma}A_{jk} - \phi_{ij}^{\sigma}h_j^M) \qquad (29)$$

$$j_i^{cR} = q_0(\chi_{ijk}^{c}A_{jk} - \phi_{ij}^{c}h_j^M) \qquad (30)$$

$$X_{ij}^{\varepsilon R} = 0 \qquad (31)$$

$$j_i^{eR} = q_0(\chi_{ijk}^{e}A_{jk} - \phi_{ij}^{e}h_j^M) \qquad (32)$$

$$\sigma_{ij}^R = -q_0(\chi_{kij}^{c}\nabla_k\mu_c + \chi_{kij}^{e}E_k + \chi_{kij}^{\sigma}\nabla_k T) \qquad (33)$$

$$Z^R = 0 \qquad (34)$$

$$Y_i^R = 0 \qquad (35)$$

$$Y_i^{\Omega R} = 0 \qquad (36)$$

$$X_i^R = q_0(\phi_{ij}^{\sigma}\nabla_j T + \phi_{ij}^{c}\nabla_j\mu_c + \phi_{ij}^{e}E_j) \qquad (37)$$

where we have disregarded gradients of the thermodynamic forces.

The material tensors χ_{ijk}^{ξ} and ϕ_{ij}^{ξ} describe purely chiral effects and contain one or two phenomenological parameters each (where $\xi \in \sigma, c, e$)

$$\chi_{ijk}^{\xi} = \chi^{\xi}(\epsilon_{ikm}n_j n_m + \epsilon_{ijm}n_k n_m) \qquad (38)$$

$$\phi_{ij}^{\xi} = \phi_1^{\xi}\delta_{ij}^{\perp} + \phi_2^{\xi}n_i n_j \qquad (39)$$

totaling 9 purely chiral reversible transport parameters.

The contributions associated with χ_{ijk}^{ξ} couple reversibly extensional flows to heat, concentration and electric currents. The symmetric second rank tensors ϕ_{ij}^{ξ} are associated with the reversible coupling of the molecular field of the magnetization, h_i^M, with the variables entropy density, concentration and electric field. All these contributions are absent in ferronematics and ferronematic gels and elastomers due to the lack of broken parity symmetry in the latter classes of materials.

3.3 Irreversible dynamics and entropy production

For ferronematic gels and elastomers we have treated the strain tensor as a conserved quantity [59]. In the present paper we incorporate not only the permanently cross-linked part of the gel, but also the effects of transient

elasticity. Therefore the part of the dissipation function associated with ψ_{ij}, the thermodynamic conjugate of the strain tensor, has two diagonal contributions, which take the form

$$R_\psi = \frac{1}{2}\gamma_{ij}\left(\nabla_k\psi_{ik}\right)\left(\nabla_l\psi_{jl}\right) + \frac{1}{2}(\tau^{-1})_{ijkl}\psi_{ij}\psi_{kl} \qquad (40)$$

where $(\tau^{-1})_{ijkl}$ has the same structure as the elastic tensor c_{ijkl} and thus five independent coefficients in a uniaxial system.

The first term is identical to the one already given in ref. [59], while the second term, which contains no gradients, describes the relaxation in the spirit of a macroscopic variable on a long, but finite time scale in the long wavelength limit [58, 81].

The other parts of the dissipation function already present for ferronematic gels and elastomers [59] as well as for ferronematic LMWs [41] have already been discussed in detail in the literature. We will not reproduce them here and only introduce some of them, when needed in the discussions in Sec. 4.

Here we focus on the dissipative part of the ferrocholesteric dynamics associated with the presence of the pseudoscalar q_0, which can be discussed most succinctly in terms of the dissipation function R [58, 76]. It is constructed in terms of the thermodynamic forces, which are either the conjugate quantities or their gradients,

$$\begin{aligned} R_L = &\ q_0\,\epsilon_{ijk}n_j h_k^n(\psi_c\nabla_i\mu_c + \psi_\sigma\nabla_i T + \psi_e E_i) \\ &+ q_0\epsilon_{ijk}n_j L_k^\perp(\psi_c^\Omega\nabla_i\mu_c + \psi_\sigma^\Omega\nabla_i T + \psi_e^\Omega E_i) \\ &+ q_0\psi_{jk}(\chi_{ijk}^{c\psi}\nabla_i\mu_c + \chi_{ijk}^{\sigma\psi}\nabla_i T + \chi_{ijk}^{e\psi}E_i) \end{aligned} \qquad (41)$$

It contains all the dissipative Lehmann-type effects relating gradients of temperature and concentration, and electric fields to director rotations (first line) and relative rotations (second line). In contrast to the static case, where there is no coupling to an electric field [78], such a contribution arises naturally in the dissipation function. On the other hand, there is no such dissipative coupling to density gradients, since there is no dissipative current associated with density (due to mass conservation). The third line describes appropriate dissipative couplings to elastic deformations, not considered before. The material tensors $\chi_{ijk}^{\xi\psi}$ with $\xi \in \{\sigma, c, e\}$ contain one dissipative transport parameter each and are of the form of eq.(38).

We stress that these dissipative contributions occur for all gels and elastomers with macroscopic chirality including cholesteric and chiral smectic gels and elastomers.

The chiral parts of the dissipative currents then read

$$j_i^{\sigma D} = -q_0(\epsilon_{ijk}n_k[\psi_\sigma h_j^n + \psi_\sigma^\Omega L_j^\perp] + \chi_{ijk}^{\sigma\psi}\psi_{jk} \quad (42)$$

$$j_i^{cD} = -q_0(\epsilon_{ijk}n_k[\psi_c h_j^n + \psi_c^\Omega L_j^\perp] + \chi_{ijk}^{c\psi}\psi_{jk} \quad (43)$$

$$j_i^{eD} = -q_0(\epsilon_{ijk}n_k[\psi_e h_j^n + \psi_e^\Omega L_j^\perp] + \chi_{ijk}^{e\psi}\psi_{jk} \quad (44)$$

$$\sigma_{ij}^D = 0 \quad (45)$$

$$Y_i^D = q_0\epsilon_{kji}n_j(\psi_c\nabla_k\mu_c + \psi_\sigma\nabla_k T + \psi_e E_k) \quad (46)$$

$$Y_i^{\Omega D} = q_0\epsilon_{kji}n_j(\psi_c^\Omega\nabla_k\mu_c + \psi_\sigma^\Omega\nabla_k T + \psi_e^\Omega E_k) \quad (47)$$

$$X_i^D = 0 \quad (48)$$

$$X_{ij}^{\epsilon D} = q_0(\chi_{kji}^{c\psi}\nabla_k\mu_c + \chi_{kji}^{\sigma\psi}\nabla_k T + \chi_{kji}^{e\psi}E_K) \quad (49)$$

totaling 9 purely chiral dissipative transport parameters, the same number as reversible ones.

As in the reversible case we have refrained from discussing effects involving gradients of the thermodynamic forces. However, if we take into account, e.g. $\nabla_i W$, then Z^D acquires chiral contributions $\sim \nabla_i h_k^n$, $\sim \nabla_j L_k^\perp$, or $\sim \nabla_j \psi_{jk}$ leading to counter terms of the form $\sim \nabla_k W$.

4 Experimental considerations

4.1 Some consequences of cross-coupling terms

The purpose of this section is to present experimentally testable predictions as a consequence of cross-couplings that are possible for ferrocholesteric LMWs and ferrocholesteric gels and elastomers. These contributions arise as a response to external fields such as flow fields or external magnetic and electric fields. Such crosscouplings can arise in the energy density and in the reversible and irreversible parts of the currents, giving rise to static and (reversible and irreversible) dynamic effects, respectively.

All the phenomenological reversible transport coefficients presented in the last section in reversible currents can assume a large range of values and different signs. (In contrast to those that have fixed values due to invariance and general symmetry constraints, such as advective and convective terms, which are made explicit in eqs. (18)–(27)). On the other hand, phenomenological crosscoupling coefficients in the energy density and in dissipative currents are bound by the requirements of positivity of energy and dissipation, respectively. Since we discuss in the following four isolated chiral cross-coupling effects, one has to make sure that additional (maybe achiral) effects do not disturb the picture given. For that reason we assume e.g. a negative dielectric anisotropy for the cases were we assume an electric field perpendicular to the nematic director.

4.2 Local piezoelectricity in ferrocholesterics

We start our analysis by inspecting the piezoelectric contribution to the generalized energy, eq.(4) of ferrocholesteric gels and elastomers (applies also to nonmagnetic, ordinary cholesteric gels and elastomers)

$$\varepsilon_p = q_0\zeta^\psi(\epsilon_{ijp}n_p n_k + \epsilon_{ikp}n_p n_j)D_i\varepsilon_{jk} \quad (50)$$

To be definite we consider the case of an electric field applied parallel to the helical axis, which we take to be oriented parallel the $\hat{z}$-axis: $E_i = E\delta_{iz}$. Then the director field, n_i, of the undeformed cholesteric helix takes the form $n_i = (\cos\tilde\varphi, \sin\tilde\varphi, 0)$ with $\tilde\varphi = \varphi(z) - \varphi_0$ containing an arbitrary initial phase φ_0, and the director field thus rotates in the $x-y$-plane perpendicular to the helical axis. Inserting these ingredients into eq.(50) we obtain for the piezoelectric contribution to the generalized energy

$$\varepsilon_p = q_0 D_z\zeta^\psi[(\varepsilon_{xx} - \varepsilon_{yy})\sin(2\tilde\varphi) - 2\varepsilon_{xy}\cos(2\tilde\varphi)] \quad (51)$$

For an external electric field, eq.(51) is minimized by period elastic deformations along the z-axis giving rise to a finite normal elastic stress difference and to elastic shear stresses, phase-shifted with respect to each other. If averaged over a large number of pitch lengths (global limit) the effect (almost) vanishes indicating that the deformations do not scale with the sample thickness. This has to be expected, since in global limit the director helix is also averaged out and cholesterics are of D_∞ symmetry that forbids piezoelectricity. For sample thicknesses that are smaller than the pitch, however, eq.(51) presents the intriguing prediction of sinusoidal variations of piezoelectric strain deformations.

4.3 An electric field induces a magnetization and a magnetic field gives a reversible electric current

To check the experimentally accessible consequences of some of the reversible dynamic contributions, we start with cross-coupling terms associated with the magnetization current and the electric current, eq.(32) and (37),

$$X_i^R = q_0\phi_{ij}^e E_j \quad\text{and}\quad j_i^{eR} = -q_0\phi_{ij}^e h_j^M \quad (52)$$

with $\phi_{ij}^e = \phi_1^e\delta_{ij}^\perp + \phi_2^e n_i n_j$, where h_i^M is the thermodynamic conjugate of the magnetization, M_i.

Assuming again an electric field along the helical axis $\hat{z}$, there is a constant reversible magnetization current $X_z^R = q_0\phi_1^e E_z$. For a sensible interpretation of this result, we have to add the (achiral) relaxation of the magnetization [59] resulting in the dynamic equation

$$\dot{M}_z + q_0\phi_1^e E_z + (1/\tau_\perp^M)M_z = 0 \quad (53)$$

with $\tau_\perp^M$ the transverse magnetization relaxation time. Thus, in the stationary limit, a finite magnetization

$$M_z = -q_0\tau_\perp^M\phi_1^e E_z \quad (54)$$

is induced by the external electric field. As a secondary effect the latter also leads to a reversible electric current $j_z^{eR} = (q_0\phi_1^e)^2\tau_\perp^M(\chi_\perp/\mu_\perp)E_z$ with the transverse magnetic susceptibility $\chi_\perp$ and $\mu_\perp = 1 + \chi_\perp$.

A kind of converse effect also exists. Considering an external magnetic field along the helical axis we get $h_z^M = -B_z$ neglecting the induced magnetization, there is a finite, constant, and reversible electric current

$$j_z^{eR} = q_0\phi_1^e B_z . \quad (55)$$

Such a connection between a vector (electric current) and an axial vector (magnetic field) is only possible in a system with broken parity (here due to pseudoscalar q_0): Under a spatial inversion (e.g. for $z \to -z$) $q_0 \to -q_0$ (while B_z is unchanged), and as a result, $j_z^{eR} \to -j_z^{eR}$, characteristic for a vector. Changing, on the other hand, the direction of time, $B_z \to -B_z$, and as a result, $j_z^{eR} \to -j_z^{eR}$, indicating that this current is reversible.

These ferroic-like effects, eqs. (54) and (55), do not require an elastic degree of freedom and are therefore present, not only in cholesteric magnetic elastomers and gels, but also in ferrocholesterics. Neither (non-magnetic) nematic elastomers and gels, nor ordinary cholesterics or cholesteric elastomers and gels can exhibit such a macroscopic behavior.

We close this subsection by pointing out that analogous effects are predicted to arise, when the electric field is replaced by temperature or concentration gradients, involving heat or concentration currents (instead of the electric current) – only the reversible transport parameters are different (ϕ_1^σ or ϕ_1^c replacing ϕ_1^e).

4.4 Flow generates reversible currents

Next we discuss reversible dynamic cross-couplings between flow on the one hand and electric fields and temperature or concentration gradients on the other. We will formulate the effects in terms of the temperature gradient and the heat current, only. The other effects are obtained using the concentration gradient or an electric field (instead of the temperature gradient) and the concentration and electric current (instead of the heat current), as well as taking the transport parameters with superscript c or e (instead of σ), respectively.

From eqs. (33) and (29) we get

$$j_i^{\sigma R} = q_0 \chi_{ijk}^\sigma A_{jk} \quad \text{and} \quad \sigma_{ij}^R = -q_0 \chi_{kij}^\sigma \nabla_k T \qquad (56)$$

with the material tensor

$$\chi_{ijk}^\sigma = \chi^\sigma (\epsilon_{ikp} n_j n_p + \epsilon_{ijp} n_k n_p) \qquad (57)$$

Assuming a simple shear flow $A_{jk} = S \delta_{jx} \delta_{ky}$ in the plane perpendicular to the helical axis, $\hat{z}$, there is a reversible heat current along the helical axis

$$j_z^{\sigma R} = -S q_0 \chi^\sigma \cos(2\tilde{\varphi}) , \qquad (58)$$

It is periodic and changes sign twice going along one pitch length resulting in local accumulation or depletion of heat. If averaged over a large number of pitch lengths the effect (almost) vanishes and for an experimental realization material with a large pitch is necessary. For a realistic comparison with appropriate experiments one has to take into account other, competing effects, e.g. the flow alignment (distorting the helical structure) and the coupling of flow with temporal changes of the strain field (requiring the use of oscillatory shear flow).

The converse effect is obtained by applying a temperature gradient along the helical axis resulting in stresses in the perpendicular plane

$$\sigma_{xx}^R = -\sigma_{yy}^R = -q_0 \nabla_z T \chi^\sigma \sin(2\tilde{\varphi}) \qquad (59)$$
$$\sigma_{yx}^R = \sigma_{xy}^R = +q_0 \nabla_z T \chi^\sigma \cos(2\tilde{\varphi}) \qquad (60)$$

that, again, are modulated sinusoidally and vanish in the global limit. Locally, the transverse shear stresses and the in-plane normal stress difference are phase shifted with respect to one another.

We note that the effects described above do not rely on the magnetic or the elastic degree of freedom and also occur in ordinary cholesterics and cholesteric gels, and in ferrocholesterics as well.

4.5 Dissipative effects of an applied magnetic field

As an example of dissipative effects that only exist in the presence of a magnetic field, we discuss the coupling between the electric (or thermal or solutal) degree of freedom with flow. From (A.17) and (A.18) we have

$$j_i^{eD} = -q_0 H_j \zeta_{ijkl}^{eA} A_{kl} \quad \text{and} \quad \sigma_{ij}^D = -q_0 H_l \zeta_{klij}^{eA} E_k \quad (61)$$

where the structure of the fourth rank tensors is given in eq.(A.13) involving in general nine independent coefficients. Similar equations are valid for the temperature and concentration current, eqs. (A.15) and (A.16).

Here we discuss the second effect for the simple case of an external magnetic field H_i along the helical axis, $\hat{z}$. In addition, we assume that the electric field (or the temperature and concentration gradient) are applied along the same direction. Then we find for the stress tensor the non-vanishing components

$$\sigma_{xx}^D + \sigma_{yy}^D = -q_0 (\zeta_1^{\xi A} + \zeta_4^{\xi A} + 2\zeta_8^{\xi A} + 2\zeta_9^{\xi A}) H_z E_z \quad (62)$$
$$\sigma_{zz}^D = -q_0 (\zeta_1^{\xi A} + 2\zeta_2^{\xi A}) H_z E_z \qquad (63)$$
$$\sigma_{xy}^D = -q_0 \left(\frac{1}{2} [\zeta_4^{\xi A} - \zeta_1^{\xi A}] + \zeta_9^{\xi A} - \zeta_8^{\xi A} \right) \sin(2\tilde{\varphi}) H_z E_z$$
$$(64)$$

The shear stresses are modulated sinusoidally along the helical axis and vanish in the global limit. However, both, the in-plane and the longitudinal compressional stresses, are constant. Note, the overall compression or dilational stress, $\sigma_{xx} + \sigma_{yy} + \sigma_{zz}$ is non-zero.

5 Summary and conclusions

Here we have derived the macroscopic dynamics for low molecular weight ferrocholesteric liquid crystals as well as for ferrocholesteric gels and elastomers. It turns out that several of the cross-coupling terms presented have also not been derived before for the usual case of low molecular weight cholesteric liquid crystals, since the symmetry based derivation of macroscopic dynamic equations for cholesterics in their local description is

also a new special case. We have analyzed in detail the influence of chirality via the pseudoscalar quantity q_0, in particular via the new dynamic variable characteristic for ferrocholesterics, the magnetization density, as well as the additional macroscopic variables characteristic of gels and elastomers, the strain field and relative rotations. Particular emphasis is laid on dynamic cross-coupling terms including contributions linear in a magnetic field. For several of the novel terms the physical consequences have been discussed. For example, we show that in a ferrocholesteric liquid crystalline material a contribution to the electric and the heat current arises, which is sensitive to the chirality in the presence of a magnetic field. Another quite remarkable effect is the prediction of electric and heat currents due to relative rotations in ferrocholesteric gels and elastomers.

Appendix A: Dynamic contributions for ferrocholesterics and ferrocholesteric gels and elastomers linear in the magnetic field

A.1 Reversible dynamics linear in a magnetic field

Using the condition $R = 0$ and general symmetry arguments, we obtain the following expressions for the chiral parts of the reversible currents in linear order in a magnetic field H_i

$$j_i^{\sigma R} = q_0(+\mu_{jik}^\sigma H_k h_j^n + \mu_{jik}^{\sigma\Omega} H_k L_j^\perp$$
$$+c_{ijkl}^\sigma H_j \psi_{kl} + \phi_{ij}^{\sigma W} H_j W) \tag{A.1}$$

$$j_i^{cR} = q_0(+\mu_{jik}^c H_k h_j^n + \mu_{jik}^{c\Omega} H_k L_j^\perp$$
$$+c_{ijkl}^c H_j \psi_{kl} + \phi_{ij}^{cW} H_j W) \tag{A.2}$$

$$X_{ij}^{\varepsilon R} = q_0 H_l(c_{klij}^\sigma \nabla_k T + c_{klij}^c \nabla_k \mu_c + c_{klij}^e E_k) \tag{A.3}$$

$$j_i^{eR} = q_0(+\mu_{jik}^e H_k h_j^n + \mu_{jik}^{e\Omega} H_k L_j^\perp$$
$$+c_{ijkl}^e H_j \psi_{kl} + \phi_{ij}^{eW} H_j W) \tag{A.4}$$

$$\sigma_{ij}^R = 0 \tag{A.5}$$

$$Z^R = q_0 H_i(\phi_{ij}^{\sigma W} \nabla_j T + \phi_{ij}^{cW} \nabla_j \mu_c + \phi_{ij}^{eW} E_j) \tag{A.6}$$

$$Y_i^R = q_0 H_k(\mu_{ijk}^\sigma \nabla_j T + \mu_{ijk}^e E_j + \mu_{ijk}^c \nabla_j \mu_c) \tag{A.7}$$

$$Y_i^{\Omega R} = q_0 H_k(\mu_{ijk}^{\sigma\Omega} \nabla_j T + \mu_{ijk}^{e\Omega} E_j + \mu_{ijk}^{c\Omega} \nabla_j \mu_c) \tag{A.8}$$

$$X_i^R = 0 \tag{A.9}$$

where we have disregarded gradients of the thermodynamic forces.

The material tensors describing chiral effects in a magnetic field, $\phi_{ij}^{\xi W}$, μ_{ijk}^ξ, $\mu_{ijk}^{\xi\Omega}$, and c_{ijkl}^ξ where $\xi \in \{\sigma, c, e\}$,

take the form with each containing two phenomenological parameters

$$\phi_{ij}^{\xi W} = \phi_1^{\xi W} \delta_{ij}^\perp + \phi_2^{\xi W} n_i n_j \tag{A.10}$$

$$\mu_{ijk}^\xi = \mu_1^\xi n_j \delta_{ik}^\perp + \mu_2^\xi n_k \delta_{ij}^\perp \tag{A.11}$$

$$\mu_{ijk}^{\xi\Omega} = \mu_1^{\xi\Omega} n_j \delta_{ik}^\perp + \mu_2^{\xi\Omega} n_k \delta_{ij}^\perp \tag{A.12}$$

or nine

$$c_{ijkl}^\xi = c_1^\xi \delta_{ij}^\perp \delta_{kl}^\perp + c_2^\xi (\delta_{ik}^\perp \delta_{jl}^\perp + \delta_{il}^\perp \delta_{jk}^\perp) + c_3^\xi n_i n_j \delta_{kl}^\perp$$
$$+c_4^\xi n_k n_l \delta_{ij}^\perp + c_5^\xi (n_i n_k \delta_{jl}^\perp + n_i n_l \delta_{jk}^\perp)$$
$$+c_6^\xi (n_j n_k \delta_{il}^\perp + n_j n_l \delta_{ik}^\perp) + c_7^\xi n_i n_j n_k n_l$$
$$+c_8^\xi n_p n_q (\varepsilon_{ikp} \varepsilon_{jlq} + \varepsilon_{ilp} \varepsilon_{jkq})$$
$$+c_9^\xi \delta_{pq}^\perp (\varepsilon_{ikp} \varepsilon_{jlq} + \varepsilon_{ilp} \varepsilon_{jkq}) \tag{A.13}$$

totaling 45 chiral-magnetic reversible transport parameters.

Therefore, in reversible dynamics, temperature gradients, concentration gradients and electric fields trigger temporal changes of the strains (c_{ijkl}^ξ), of the degree of nematic order ($\phi_{ij}^{\xi W}$), of the relative rotations ($\mu_{ijk}^{\xi\Omega}$), as well as director rotations (μ_{ijk}^ξ). Vice versa, there are counter effects that guarantee zero entropy production, in particular there are temperature, concentration and electric currents induced by elastic stresses (c_{ijkl}^ξ), by 'molecular' fields of the nematic order ($\phi_{ij}^{\xi W}$), relative rotations ($\mu_{ijk}^{\xi\Omega}$), and the director (μ_{ijk}^ξ).

A.2 Irreversible dynamics and entropy production linear in a magnetic field

In the presence of an external magnetic field there are additional dissipative coupling terms characteristic for ferrocholesterics and ferrocholesteric gels and elastomers, which are linear in H_i

$$R_H = q_0 A_{kl} H_j(\zeta_{ijkl}^{cA} \nabla_i \mu_c + \zeta_{ijkl}^{\sigma A} \nabla_i T + \zeta_{ijkl}^{eA} E_i)$$
$$+ q_0 h_k^M H_j(\chi_{ijk}^{cM} \nabla_i \mu_c + \chi_{ijk}^{\sigma M} \nabla_i T + \chi_{ijk}^{EM} E_i) \tag{A.14}$$

relating gradients of temperature and concentration, and electric fields to flow (first line) and to rotations of the magnetization (second line). They require the simultaneous presence of macroscopic chirality and a magnetic field and have apparently not been considered before. Appropriate couplings to the nematic order parameter (involving W) are not possible due to the lack of any rank-1 tensor that is even under the n_i to $-n_i$ invariance. The property tensors $\chi_{ijk}^{\xi M}$ and $\zeta_{ijkl}^{\xi A}$, with $\xi \in \{\sigma, c, e\}$, take the form of eqs. (7) and (A.13), respectively.

The chiral parts of the dissipative currents then read

$$j_i^{\sigma D} = -q_0 H_j [\zeta_{ijkl}^{\sigma A} A_{kl} + \chi_{ijk}^{\sigma M} h_k^M] \tag{A.15}$$

$$j_i^{cD} = -q_0 H_j [\zeta_{ijkl}^{cA} A_{kl} + \chi_{ijk}^{cM} h_k^M] \tag{A.16}$$

$$j_i^{eD} = -q_0 H_j [\zeta_{ijkl}^{eA} A_{kl} + \chi_{ijk}^{eM} h_k^M] \tag{A.17}$$

$$\sigma_{ij}^D = -q_0 H_l (\zeta_{klij}^{cA} \nabla_k \mu_c + \zeta_{klij}^{\sigma A} \nabla_k T + \zeta_{klij}^{eA} E_k) \tag{A.18}$$

$$Y_i^D = 0 \tag{A.19}$$

$$Y_i^{\Omega D} = 0 \tag{A.20}$$

$$X_i^D = q_0 H_j (\chi_{kji}^{cM} \nabla_k \mu_c + \chi_{kji}^{\sigma M} \nabla_k T + \chi_{kji}^{eM} E_K) \tag{A.21}$$

$$X_{ij}^{\varepsilon D} = 0 \tag{A.22}$$

totaling 39 chiral-magnetic dissipative transport parameters.

As in the reversible case we have refrained from discussing effects involving gradients of the thermodynamic forces. However, if we take into account, e.g. $\nabla_i W$, then Z^D acquires chiral magnetic contributions $\sim \nabla_k A_{ij}$ and $\sim \nabla_k h_i^M$, leading to counter terms the form $\sim \nabla_k W$.

Appendix B: Dynamic contributions for achiral ferronematics and ferronematic gels and elastomers linear in the magnetic field

Here we summarize dissipative and reversible dynamic contributions for ferronematics and ferromagnetic gels and elastomers which are linear in a magnetic field and which arise already without chirality. The dissipative contributions as they are conveniently incorporated into the dissipation function take the form

$$\begin{aligned} R_H^{ach} = {} & A_{ij} H_k (\lambda_{ijkl}^{Dn} h_l^n + \lambda_{ijkl}^{D\Omega} L_l^\perp) \\ & + H_i A_{jk} (\lambda_{ijk}^{DW} W + \lambda_{ijklm}^{D\psi} \psi_{lm}) \\ & + H_i h_j^M (\psi_{ijk}^{Dn} h_k^n + \psi_{ijk}^{D\Omega} L_k^\perp + \psi_{ij}^{DW} W + \psi_{ijkl}^{D\psi} \psi_{kl}) \end{aligned} \tag{B.1}$$

where the second rank material tensor is of the standard uniaxial form, eq. (39), the rank-3 tensor λ_{ijk}^{DW} is symmetric in j and k and as in eq. (38), while $\psi_{ijk}^{D\alpha}$ for $\alpha \in \{n, \Omega\}$ is transverse in the last index and odd in the number of n's and given by

$$\psi_{ijk}^{D\alpha} = \psi_1^{D\alpha} n_i \delta_{jk}^\perp + \psi_2^{D\alpha} n_j \delta_{ik}^\perp \tag{B.2}$$

The rank-4 tensor $\psi_{ijkl}^{D\psi}$ is symmetric in two indices and is given by eq.(A.13), while for $\lambda_{ijkl}^{D\alpha}$ we have

$$\begin{aligned} \lambda_{ijkl}^{D\alpha} = {} & \lambda_1^{D\alpha} n_i n_j n_p \varepsilon_{klp} + \lambda_2^{D\alpha} \delta_{ij}^\perp \varepsilon_{klp} n_p \\ & + \lambda_3^{D\alpha} (n_i n_k n_p \varepsilon_{jlp} + n_j n_k n_p \varepsilon_{ilp}) \\ & + \lambda_4^{D\alpha} (\delta_{il}^\perp n_p \varepsilon_{jkp} + \delta_{jl}^\perp n_p \varepsilon_{ikp}) \\ & + \lambda_5^{D\alpha} (\delta_{ik}^\perp n_p \varepsilon_{jlp} + \delta_{jk}^\perp n_p \varepsilon_{ilp}) \end{aligned} \tag{B.3}$$

We refrain from writing down the rather cumbersome structure of the fifth rank tensor $\lambda_{ijklm}^{D\psi}$ in detail here.

We note that, except for the coupling between extensional flow and the nematic molecular field, λ_{ijkl}^{Dn}, which has been given in ref. [41], all other coupling terms are presented here for the first time. The third line of eq.(B.1) shows in particular that the molecular field, h_i^M, associated with the magnetization, can couple dissipatively to the molecular field of the director as well as to relative rotations (via $L_i^\perp$).

If we make use of the symmetry arguments outlined in the main part of this paper and use the condition $R = 0$ we obtain the following phenomenological expressions for the reversible currents up to linear order in the thermodynamic forces and linear in the magnetic field, H_i

$$j_i^{\sigma R} = \psi_{ijk}^{\sigma\sigma} H_j \nabla_k T + \psi_{ijk}^{\sigma c} H_j \nabla_k \mu_c + \psi_{ijk}^{\sigma E} H_j E_k \tag{B.4}$$

$$j_i^{cR} = -\psi_{kji}^{\sigma c} H_j \nabla_k T + \psi_{ijk}^{cc} H_j \nabla_k \mu_c + \psi_{ijk}^{cE} H_j E_k \tag{B.5}$$

$$\begin{aligned} X_{ij}^{\varepsilon R} = {} & \tau_{ijklm}^{\psi\psi} H_m \psi_{kl} + \psi_{ijk}^{\psi W} H_k W \\ & + \psi_{ijkl}^{\psi n} H_k h_l^n + \psi_{ijkl}^{\psi\Omega} H_k L_l^\perp \end{aligned} \tag{B.6}$$

$$j_i^{ER} = -\psi_{kji}^{\sigma E} H_j \nabla_k T - \psi_{kji}^{cE} H_j \nabla_k \mu_c + \psi_{ijk}^{EE} H_j E_k \tag{B.7}$$

$$\sigma_{ij}^R = -\nu_{ijklm}^R H_m A_{kl} - \psi_{ijkl}^{AM} H_k h_l^M \tag{B.8}$$

$$Z^R = -\psi_{ijk}^{\psi W} H_k \psi_{ij} + \chi_W \delta_{ij}^\perp H_j h_i^n + \chi_\Omega \delta_{ij}^\perp H_j L_i^\perp \tag{B.9}$$

$$\begin{aligned} Y_i^R = {} & (\gamma^{-1})_{ijk}^R h_j^n H_k + \tilde{\Theta}_{ijk} L_j^\perp H_k \\ & - \chi_W \delta_{ij}^\perp H_j W + \psi_{klji}^{\psi n} H_j \psi_{kl} \end{aligned} \tag{B.10}$$

$$\begin{aligned} Y_i^{\Omega R} = {} & \Theta_{ijk} L_j^\perp H_k - \tilde{\Theta}_{jik} h_j^n H_k \\ & - \chi_\Omega \delta_{ij}^\perp H_j W + \psi_{klji}^{\psi\Omega} H_j \psi_{kl} \end{aligned} \tag{B.11}$$

$$X_i^R = \psi_{ijk}^{MM} H_j h_k^M + \psi_{klji}^{AM} H_j A_{kl} \tag{B.12}$$

where the counter-terms guarantee vanishing entropy production. For the self-coupling terms that do not have counter-terms, the contribution to R has to vanish identically. We have for $(\gamma^{-1})_{ijk}$, $\tilde{\Theta}_{ijk}$ and Θ_{ijk}

$$\begin{aligned} \theta_{ijk} = {} & \theta_1 (\varepsilon_{ijk} + n_p n_i \varepsilon_{pkj} - n_p n_j \varepsilon_{pki}) \\ & + \theta_2 \varepsilon_{pij} n_p n_k \end{aligned} \tag{B.13}$$

For the third rank tensors $\psi_{ijk}^{\alpha\beta}$ with $\{\alpha, \beta\} \in \{\sigma, c, E, M\}$ we find the same form as eq. (7) for $\alpha \neq \beta$

$$\begin{aligned} \psi_{ijk}^{\alpha\beta} = {} & \psi_1^{\alpha\beta} \epsilon_{ijk} + \psi_2^{\alpha\beta} \epsilon_{ijp} n_p n_k \\ & + \psi_3^{\alpha\beta} \epsilon_{ipk} n_p n_j + \psi_4^{\alpha\beta} \epsilon_{pjk} n_p n_i \end{aligned} \tag{B.14}$$

while for $\alpha = \beta$ the additional condition $\psi_2^{\alpha\alpha} = \psi_4^{\alpha\alpha}$, leading to $\psi_{ijk}^{\alpha\alpha} = -\psi_{kji}^{\alpha\alpha}$, is required to give $R = 0$.

The tensor $\psi_{ijk}^{\psi W}$ is symmetric in i and j and of the form

$$\psi_{ijk}^{\psi W} = \psi^{\psi W} (\epsilon_{kjp} n_p n_i + \epsilon_{kip} n_p n_j) \tag{B.15}$$

The fourth order tensors are symmetric in the first pair of indices and either even in n, as is ψ_{ijkl}^{AM} or odd in n and

transverse in l, as are $\psi_{ijkl}^{\psi n}$ and $\psi_{ijkl}^{\psi \Omega}$. The former is given by Eq. (A.13), and the latter by (B.3), containing nine and five coefficients, respectively.

The expression for $\nu_{ijklm}^R H_m$ as well as its symmetry properties have been studied in detail in ref. [41]. The corresponding analysis applies equally well to $\tau_{ijklm}^{\psi\psi} H_m$.

For ferronematics the coupling terms between heat and concentration currents on the one hand and temperature and chemical potential gradients on the other have been given in ref. [41]. This also applies to the coupling terms between extensional flow and the stress tensor and between the director quasi-current and the molecular field of the director. All contribution involving relative rotations, the magnetization, the order parameter modulus and the electric field and their respective thermodynamic forces and (quasi-)currents are presented here in eqs.(B.4) - (B.12) for the first time.

Appendix C: Selected aspects of the rheological behavior of ferrocholesterics and ferrocholesteric gels and elastomers

In the case of ferronematic materials (low molecular weight as well as gels and elastomers) one has been able to find explicitly and quite generally a stationary solution under simple shear (Sec. 4.2 of ref. [59]). This is no longer possible for the case of ferrocholesteric materials due to the intricate coupling between all macroscopic variables. We expect, however, that future measurements of the rheological properties of ferrocholesteric materials can serve as a guide for making suitable simplifying assumptions to the general problem so that explicit expressions for the complex dynamic shear modulus, $G^*(\mathbf{k}, \omega) = G'(\mathbf{k}, \omega) + iG''(\mathbf{k}, \omega)$, as well as for the complex dynamic Young modulus, $E^*(\mathbf{k}, \omega) = E'(\mathbf{k}, \omega) + iE''(\mathbf{k}, \omega)$, can be extracted from the general macroscopic basic equations.

In this Appendix we discuss how the broken mirror symmetry modifies the elastic mechanical stress, ψ_{ij}, the stress tensor, σ_{ij}, and X_{ij}^ϵ that describes the dynamic changes of the strain tensor – the quantities that come into play naturally during measurements of G^* and E^*. These chiral effects are proportional to q_0 and their experimental relevance increases with q_0. For samples of thickness L with $Lq_0 \gg 1$, there are, however, very many pitch lengths present. Those chiral effects that are periodic with the helix are averaged out in that case. Only for very thin samples with $Lq_0 \sim 1$, they survive, but are difficult to detect, since the situation is almost nematic-like. Therefore, most important experimentally are those chiral effects that do not vanish, when averaged over many pitch lengths.

The chiral contributions to the static stress ψ_{ij}, eq.(13), only arise for chiral gels and elastomers. In sec. 4.2 we have discussed in detail the piezoelectric (second) contribution in eq.(13), and found that it vanishes when averaged over many pitch lengths (in thick samples). In contrast, the first term, describing the influence of changes in the helical pitch, can be expected to contribute for all

sample thicknesses to elongational flows and deformations, since

$$\psi_{xx} + \psi_{yy} = q_0(\tau_1^\varepsilon + \tau_2^\varepsilon)\nabla_z(\delta\tilde{\phi}) \qquad (C.1)$$
$$\psi_{zz} = q_0\tau_2^\varepsilon \nabla_z(\delta\tilde{\phi}) \qquad (C.2)$$

while the shear components either vanish identically or when averaged over many pitch lengths

$$\psi_{xy} = q_0(\tau_1^\varepsilon - \tau_2^\varepsilon)\sin(2\tilde{\varphi})\nabla_z(\delta\tilde{\phi}) \qquad (C.3)$$
$$\psi_{xz} = \psi_{yz} = 0 \qquad (C.4)$$

For the case without an external magnetic field there are only reversible contributions to the stress tensor σ_{ij}^R, given by eq.(33). As discussed in Sec. 4.4 all contributions, either due to a temperature, a chemical potential gradient, or an electric field, applied along the helical axis, are periodic and vanish when averaged over many pitch lengths.

Dissipative contributions to the stress tensor, σ_{ij}^D, are possible in the presence of a magnetic field, eq. (A.18). As discussed in section 4.5 only the diagonal elements of the stress tensor survive the averaging process, cf. eqs. (62) and (63).

The dynamic contributions to X_{ij}^ϵ behave rather opposite: The dissipative contributions (without a field), eq.(48), all vanish when averaged, while the reversible one, (in the presence of a magnetic field), eq.(A.3), have the same structure as eqs.(C.1)-(C.4) and only the diagonal elements are non-zero when averaged

$$X_{xx}^\epsilon + X_{yy}^\epsilon = q_0(c_1^\sigma + c_4^\sigma + 2c_8^\sigma + 2c_9^\sigma)\nabla_z T \quad (C.5)$$
$$X_{zz}^\epsilon = q_0(c_1^\sigma + 2c_2^\sigma)\nabla_z T \qquad (C.6)$$

here written down for the case of a temperature gradient.

These results, together with the appropriate counter terms, give us some hints about the role of the chiral contributions to G^* and E^*. First, shear excitations are only weakly influenced, since the chiral contributions vanish when averaged over many pitch lengths. On the other hand, sound-like excitations do get (robust) chiral contributions, both in the real and the imaginary part of E^*. The static contributions, eq.(C.1) and (C.2), show that changing the effective pitch, e.g. by rotating one of the horizontal boundary plates (assuming strong nematic anchoring), leads to residual elastic stresses that change effectively the elastic modulus and therefore $E'(\boldsymbol{k}, \omega)$.

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