ELECTROHYDRODYNAMICS

OF NEMATIC LIQUID CRYSTALLINE ELASTOMERS

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We discuss the macroscopic behaviour of nematic liquid crystalline side-chain elastomers. These systems are particularly interesting from a macroscopic point of view, since they allow the occurence of relative rotations between the nematic director and the permanent elastomeric network. Both, the purely hydrodynamic as well as the electrohydrodynamic equations are presented. The differences and the similarities to the macroscopic behaviour of nematic liquid crystalline side-chain polymers are clarified.

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1. Introduction

About fifteen years ago [1] liquid crystalline side-chain polymers have first been synthesized. Since then their synthesis and the study of their properties has become a rapidly growing field of polymer science (compare for example refs. 2 and 3 for reviews). Although liquid crystalline side-chain elastomers have been synthesized only three years [4] after the first report of liquid crystalline side-chain polymers, the number of investigations on liquid crystalline elastomers was much smaller for many years [5-17], partly because it was difficult to generate samples with homogeneous cross-linking density and reproducible spatial homogeneity. The interest increased recently [18-31], especially because it became possible to generate 'single crystal' liquid crystalline elastomers either by two cross-linking steps [26] or by orienting a nematic polymer in a magnetic field at the beginning of the cross-linking reaction [27] and since cholesteric and chiral smectic elastomers show interesting electromechanical effects [18,20-22,30,31].

While there has been a considerable amount of work on the nematic - isotropic transition in nematic side-chain and main-chain elastomers [4,7,8,10,11,15-17,28,32,33], comparatively few articles have addressed macroscopic mechanical and electromechanical properties of nematic elastomers [5,12,14,19,29,34].

In this paper we will discuss theoretically the static and the dynamic macroscopic and electrohydrodynamic properties of nematic side-chain elastomers. In this general area the only piece of work we could find was the seminal contribution by de Gennes [34], who introduced the concept of relative rotations in nematic gels and who presented contributions to the generalized free energy coupling director rotations to the permanent elastomeric network. Here we will generalize ref.[34] by allowing for couplings to external magnetic and electric fields. In addition we will present dynamic equations for nematic side-chain elastomers, assuming that the cross-linking density is sufficiently low so that both, the nematic director and the strain tensor are legitimate hydrodynamic variables [35-37]. As the cross-linking density is increased, the average distance between neighbouring

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reticulation points shrinks and the use of a director field as an independent macroscopic variable is no longer justified. At sufficiently high cross-linking densities a solid-like hard rubber with no additional director-type orientational degrees of freedom results.

After a general introduction to the method, which we use to set up the macroscopic dynamic description of nematic liquid-crystalline side-chain elastomers (sec. 2), we introduce the hydrostatic and static electromechanical effects in liquid crystalline elastomeric phases in sec.3. In section 4 we give the dynamic and electrohydrodynamic equations and in section 5 we critically discuss the limitations of the approach and give a perspective for further generalizations.

2. Hydrodynamic and Electrohydrodynamic Equations

The derivation of hydrodynamic equations [35-37] is a well established procedure, which yields balance equations describing the macroscopic response of a system in the long wavelength, low frequency limit. Here long wavelengths means length scales large compared to all microscopic lengths amd low frequencies refers to frequencies small compared to all collisional frequencies.

In simple liquids and mixtures of miscible liquids [35] all hydrodynamic equations are a consequence of conservation laws, namely of those for particle number, linear momentum, energy and concentration (the last in mixtures). In liquid crystalline phases [38], additional quantities must be taken into account to characterize the low frequency, long wavelength behaviour of these systems [36,37]. These additional hydrodynamic variables are not related to conservation laws, but they are associated with spontaneously broken continuous symmetries. For example, in low moelcular weight uniaxial nematic liquid crystals [38] there is a preferred direction in space (characterized by the director $\hat{\mathbf{n}}$, a unit vector that does not distinguish between head and tail), although the Hamiltonian of the system is isotropic. Positional order in these systems is still short ranged just as for an isotropic liquid. It turns out [36-38], that the deviations δn_i from the preferred direction

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$\hat{\mathbf{n}}$ are the hydrodynamic variables [36,37] associated with the spontaneously broken rotational symmetry of nematic liquid crystals. As any true hydrodynamic variable, they show only a slow response ($\omega \to 0$) in the homogeneous limit ($k \to 0$) with diffusion as the mechanism of dissipation. In a crystal, where one has long range positional order, the displacement vector $\mathbf{u}$ becomes the variable associated with the broken translational symmetry. This additional broken symmetry gives rise to the transverse acoustic phonons in crystals. Similarly, in the field of liquid crystals, the displacement of smectic layers gives rise to the second sound in smectic A and C [36].

For nematic side-chain elastomers made by cross-linking nematic side-chain polymers, undoubtedly the displacement field must be kept as a hydrodynamic variable, since the permanent network implies a finite shear modulus at vanishing frequency, a characteristic feature of solids [36,37]. In order for the director variations δn_i to be good hydrodynamic variables, it is important that the average distance between cross-linking points is sufficiently large so that nematic order can be sensibly defined, a point already made in ref.[34] and called weak nematic gels there. As the cross-linking density is increased, it will no longer be possible to define a director field and eventually, further increasing the cross-linking density, an ordinary strongly cross-linked rubber results.

Accordingly we focus here on nematic elastomers, which are weakly cross-linked and for which therefore the director variations δn_i can be viewed as deviations from a nematic director $\hat{\mathbf{n}}$. To take into account elastic effects, we will expand all potentials, thermodynamic forces and the entropy production into powers of the strain tensor $\epsilon_{ij} = \frac{1}{2}(\nabla_i u_j + \nabla_j u_i)$. This concept has turned out to be a useful one [15,28] at least in the isotropic phase above the isotropic - nematic transition for elongations of the elastomers by up to about 50% of its original length. It goes without saying that such an expansion is only valid for sufficiently small strains.

In addition to considering the strain tensor and spatial variations of the director field $\hat{\mathbf{n}}$, it has been pointed out by de Gennes [34], that it is also important to keep relative

rotations of the director with respect to the elastomeric network. In ordinary solids local rotations Ω_{ij} (where in linearized theory $\Omega_{ij} = \frac{1}{2}(\nabla_i u_j - \nabla_j u_i)$) do not contribute to the macroscopic behaviour, since the thermodynamic potentials must be invariant under rigid rotations. Thus only gradients of local rotations are allowed to enter in the corresponding expressions for ordinary solids, which are usually discarded, since they represent higher order gradient terms. In nematic elastomers, however, these local rotations must be split into two groups. First there are the rotations around the preferred direction $\hat{\mathbf{n}}$ ($\Omega^{\|} = n_i \epsilon_{ijk} \Omega_{jk}$). These do not involve, however, rotations relative to $\hat{\mathbf{n}}$ and are therefore decoupled from the hydrodynamic variables. More importantly, however, there are two rotations orthogonal to the first one ($\Omega_i^{\perp} = n_j \Omega_{ij}$ with $n_i \Omega_i^{\perp} = 0$). These can give rise to *relative* rotations ($\tilde{\Omega}_i = \delta n_i - \Omega_i^{\perp}$ with $\mathbf{n} \cdot \delta \mathbf{n} = 0$) and will play an important role in the following sections, as they are used to construct contributions to the thermodynamic potentials etc., a possibility that was first recognized in ref. [34]. We note that, by construction, $\tilde{\Omega}_i$ has the same transformation behaviour under $\hat{\mathbf{n}} \to -\hat{\mathbf{n}}$ as the director itself.

We note in passing that the use of relative rotations is rather similar to the use of relative translations of two displacement vectors along the same direction, a concept that plays an important role in the characterization of the macroscopic behaviour of incommensurate solid systems [39]. We would like to stress that the relative rotations introduced above are not linked to any additional independent hydrodynamic degrees of freedom - in addition to the director deviations δn_i and the displacement field u_i - since these relative rotations are neither connected to a conserved quantity nor to a spontaneously broken continuous symmetry. Thus they can be expected to relax at best on a long, but finite timescale and belong to the class of so-called macroscopic variables (compare the following discussion).

As truly hydrodynamic variables we have therefore the conserved quantities density ρ, density of linear momentum $\mathbf{g}$ and energy density ϵ along with the variables associated

with the spontaneously broken continuous symmetries, namely the displacement field $\mathbf{u}$ (associated with the broken translational symmetry) and the director variations δn_i (associated with the broken rotational symmetry and where we have assumed that a director field can be sensibly defined via a large enough reticulation distance and a sufficiently small coupling via flexible spacers between the mesogenic units and the elastomeric backbone). For mixtures this list has to be extended by the concentration variable c.

In addition to these truly hydrodynamic variables we will also consider in the following the influence of external electric fields and we will therefore keep the local electric field $\mathbf{E}$ (or, equivalently, the macroscopic polarization $\mathbf{P}$ or the electric displacement $\mathbf{D} \equiv \mathbf{E} + 4\pi\mathbf{P}$) as a macroscopic variable, which includes the electric charge conservation law (compare, for example, ref.[40] for a detailed exposition on the incorporation of the effects of electric fields into nematodynamics). In the main text we will use $\mathbf{D}$ as variable, but in the Appendix we will list the equivalent formulas using $\mathbf{E}$ as the variable. Furthermore we will keep in our list of macroscopic variables the relative rotations already discussed above as well as variations of the modulus of the nematic order parameter to allow for the fact that backbone fluctuations not only couple to orientational fluctuations of the director but also influence the degree of nematic ordering. Thus, order fluctuations δS may be slowed down considerably in elastomeric systems and are then relevant for a macroscopic description (in contrast to low molecular weight systems, where δS is a fast variable usually not kept in a macroscopic description except near the nematic to isotropic phase transition).

In closing this section we would like to note that we will not consider throughout the bulk part of this paper the effect that chain entanglements give rise to an additional (in addition to the permanent network) transient network above a certain characteristic frequency as it is already the case for polymer melts and solutions, regardless whether they are isotropic or liquid crystalline [41-47]). This issue will be addressed briefly in section 6 in order not to overload the following presentation.

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3. Static Behaviour of Nematic Elastomers

To derive the complete set of hydrodynamic and electrohydrodynamic equations, we make use of irreversible thermodynamics and general symmetry arguments including the transformation behaviour of all thermodynamic quantities under time reversal, parity, translations, rigid rotations, Galilean invariance [35-37, 48] and - for nematic side-chain elastomers - under the replacement $\hat{\mathbf{n}} \rightarrow -\hat{\mathbf{n}}$.

The starting point of the general procedure is the local formulation of the first law of thermodynamics, the Gibbs-Duhem relation [48] relating changes of the entropy density σ to the other macroscopic variables. For nematic elastomers we have, incorporating all the macroscopic variables listed above and assuming that all other (microscopic) variables are in thermodynamic equilibrium on the relevant time and length scales

$$T d\sigma = d\epsilon - \mu d\rho - \mu_c dc - v_i dg_i - W dS + h_i dn_i - \psi_{ij} d\epsilon_{ij} - L_i^\perp d\tilde{\Omega}_i - E_i dD_i \qquad (3.1)$$

Summation over repeated indices is implied in eq.(3.1) and all the following equations unless specified otherwise. In the absence of any external fields the direction of $\hat{\mathbf{n}}$ is arbitrary (because rotational symmetry is broken *spontaneously*) and homogeneous rotations dn_i (with $n_i dn_i = 0$) do not cost energy. In that case $h_i dn_i$ has to be replaced by $-\phi_{ij} d(\nabla_j n_i)$. The thermodynamic conjugate quantities chemical potential μ, relative chemical potential of the mixture μ_c, velocity v_i, polarization P_i, temperature T, nematic molecular field h_i, elastic stress ψ_{ij}, 'modulus field' W and relative molecular field $L_i^\perp$ are defined via eq.(3.1).

To determine these thermodynamic forces and thus the static properties of nematic elastomers one expands the generalized energy density ϵ into the variables and then obtains the forces by taking a variational derivative with respect to one variable while keeping all other variables at a fixed value.

We find for the generalized energy for nematic elastomers $E = E_0 + \int d\tau \epsilon$ up to quadratic order in the variables, where E_0 contains all the terms already present in miscible

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mixtures of two simple liquids

$$\epsilon = \frac{1}{2}c_{ijkl}\epsilon_{ij}\epsilon_{kl} + (\chi_{ij}^{\rho}\delta\rho + \chi_{ij}^{\sigma}\delta\sigma + \chi_{ij}^{c}\delta c + \chi_{ij}^{S}\delta S)\epsilon_{ij}$$
$$+ \frac{1}{2}K_{ijkl}(\nabla_j n_i)(\nabla_l n_k) + \frac{a}{2}(\delta S)^2 + (b^{\sigma}\delta\sigma + b^{\rho}\delta\rho + b^c\delta c)\delta S$$
$$+ \frac{1}{2}(\frac{1}{\epsilon^E})_{ij}D_iD_j + \frac{1}{2}\chi_{ij}H_iH_j + \frac{1}{2}D_1\tilde{\Omega}_i\tilde{\Omega}_i + D_2\tilde{\Omega}_i\epsilon_{jk}n_j\delta_{ik}^{\perp} \qquad (3.2)$$
$$+ e_{ijk}(\nabla_i n_j)D_k + e_{ij}^{S}(\nabla_i\delta S)D_j + M_{ijk}(\nabla_i n_j)(\nabla_k\delta S)$$
$$+ \chi_{ijkl}^{N}(\nabla_i D_j)\epsilon_{kl} + \gamma_{ijk}^{N}\tilde{\Omega}_k\nabla_i D_j$$

Certain additional electrostrictive and magnetostrictive effects that couple the fields (quadratically) to the strain and to the relative rotations will be discussed in the Appendix. In eq.(3.2) the elastic tensor has the form familiar from uniaxial solids [36, 49, 50]

$$c_{ijkl} = c_1\delta_{ij}^{\perp}\delta_{kl}^{\perp} + c_2(\delta_{ik}^{\perp}\delta_{jl}^{\perp} + \delta_{il}^{\perp}\delta_{jk}^{\perp}) + c_3 n_i n_j n_k n_l$$
$$+ c_4(\delta_{ij}^{\perp}n_k n_l + \delta_{kl}^{\perp}n_i n_j) + c_5(\delta_{ik}^{\perp}n_j n_l + \delta_{il}^{\perp}n_k n_j + \delta_{jk}^{\perp}n_i n_l + \delta_{jl}^{\perp}n_k n_i) \qquad (3.3)$$

with $\delta_{ij}^{\perp} = \delta_{ij} - n_i n_j$. The tensor K_{ijkl} is the usual Frank curvature elastic tensor [36-38] $K_{ijkl} = K_1\delta_{ij}^{\perp}\delta_{kl}^{\perp} + K_2 n_p\epsilon_{pij}n_q\epsilon_{qkl} + K_3\delta_{ik}^{\perp}n_j n_l$. The static susceptibility tensors χ_{ij}^{ρ}, χ_{ij}^{σ}, χ_{ij}^{c} and χ_{ij}^{S} describing static cross-couplings of density, entropy density, concentration and order parameter to the diagonal part of the strain tensor as well as the (reciprocal) dielectric and the diamagnetic susceptibility tensors $(1/\epsilon^E)_{ij}$ and χ_{ij} take the uniaxial form $\chi_{ij} = \chi_{\parallel}n_i n_j + \chi_{\perp}\delta_{ij}^{\perp}$. The contributions proportional to D_1 and D_2 containing relative rotations betwen the elastomeric network and the director field are the ones that have been given already by de Gennes in ref.[34]. The flexoelectric tensor is generally written as [33] $e_{ijk} = e_1\delta_{ij}^{\perp}n_k + e_3\delta_{jk}^{\perp}n_i$ The contribution $\propto e_{ij}^{S}$, which couples gradients of the modulus to the electric field, reads $e_{ij}^{S} = e_{\perp}^{S}\delta_{ij}^{\perp} + e_{\parallel}^{S}n_i n_j$ and the coupling M_{ijk} between gradients of the director and of the modulus [51] brings along one extra coefficient $M_{ijk} = M(n_i\delta_{jk}^{\perp} + n_k\delta_{ij}^{\perp})$. The contribution $\propto \chi_{ijkl}^{N}$ represents a coupling between strains and spatially inhomogeneous electric fields. This is a term not usually considered in solids, but it might play a more important role in nematic elastomers, since these materials are

frequently only weakly cross-linked and can also have highly polarizable groups in their mesogenic units.

The tensor χ_{ijkl}^N assumes a slightly more complicated form [50] than eq.(3.3) since it lacks the $\chi_{ijkl}^N = \chi_{klij}^N$ symmetry, but it fulfils $\epsilon_{jp}^E \chi_{ipkl}^N = \epsilon_{ip}^E \chi_{jpkl}^N$ provided we make use of $curl\mathbf{E} = 0$ (cf. App.). There is a corresponding coupling between gradients of electric fields and relative rotations, where the tensor assumes the form $\gamma_{ijk}^N = \gamma_1 n_i \delta_{jk}^\perp + \gamma_2 n_j \delta_{ik}^\perp$, containing only one parameter, if $curl\,\mathbf{E} = 0$ is used, since then $\gamma_1 \epsilon_\parallel^E = \gamma_2 \epsilon_\perp^E$, (cf. App.). The new terms ($\sim \gamma_{ijk}^N$) are only possible, however, in systems that allow for the possibility of relative rotations.

For the generalized forces $L_i^\perp$, ψ_{ij}, h_i, W and E_i we find from eq.(3.2) by taking the variational derivatives with respect to $\tilde{\Omega}_i$, ϵ_{ij}, n_i, S and D_i while keeping all other variables constant

$$L_i^\perp = \frac{\delta E}{\delta \tilde{\Omega}_i}|_{...} = D_1 \tilde{\Omega}_i + D_2 \epsilon_{jk} n_j \delta_{ik}^\perp + \gamma_{jki}^N \nabla_j D_k \tag{3.4}$$

$$\psi_{ij} = \frac{\delta E}{\delta \epsilon_{ij}}|_{...} = c_{ijkl}\epsilon_{kl} + \chi_{ij}^\rho \delta\rho + \chi_{ij}^\sigma \delta\sigma + \chi_{ij}^c \delta c + \chi_{ij}^S \delta S$$
$$+ \frac{1}{2} D_2 (\tilde{\Omega}_i n_j + \tilde{\Omega}_j n_i) + \chi_{ijkl}^N \nabla_k D_l \tag{3.5}$$

$$h_k \delta_{ik}^\perp = -\delta_{ik}^\perp \frac{\delta E}{\delta n_k}|_{...} = h_i^{(2)} + \delta_{ip}^\perp \nabla_j \Big(K_{pjkl} \nabla_l n_k + e_{jpk} D_k + M_{jpk} \nabla_k \delta S \Big) \tag{3.6}$$

$$W = \frac{\delta E}{\delta S}|_{...} = a\delta S + \chi_{ij}^S \epsilon_{ij} + b^\sigma \delta\sigma + b^\rho \delta\rho + b^c \delta c - \nabla_k (M_{ijk}\nabla_i n_j) - \nabla_j (e_{ij}^S D_i) \tag{3.7}$$

$$E_i = \frac{\delta E}{\delta D_i}|_{...} = (\frac{1}{\epsilon^E})_{ij} D_j + e_{jki}\nabla_j n_k + e_{ij}^S \nabla_j \delta S - \nabla_j (\chi_{jikl}^N \epsilon_{kl}) - \nabla_j (\gamma_{jik}^N \tilde{\Omega}_k) \tag{3.8}$$

For the remaining thermodynamic conjugates μ, μ_c and v_i one can proceed in the same way. In eq.(3.6) the non-divergence part of h_i, $h_i^{(2)}$, contains the orientational effects that fix the direction of $\hat{\mathbf{n}}$ in external fields, i.e. the dielectric and diamagnetic anisotropy effect ($\propto -(1/\epsilon_a)\delta_{ij}^\perp n_k D_j D_k - \chi_a \delta_{ij}^\perp n_k H_j H_k$) as well as various other nonlinear orienting effects on $\hat{\mathbf{n}}$ due to e.g. strains ($\propto -\epsilon_{pj}\epsilon_{kl}(\partial/\partial n_i)c_{pjkl}$) or due to the combined action of strains, fields, relative rotations, gradients of S, gradients of $\mathbf{n}$ and gradients of the other variables. Since strains can become rather large in elastomers, the orienting effect quadratic in the

strains can be expected to be important.

The static susceptibilities contained in eqs.(3.4) - (3.8) are taken at constant $\mathbf{D}$; their relations to those at constant $\mathbf{E}$ are given in the Appendix.

4. Dynamic Behaviour of Nematic Elastomers

To derive the complete set of dynamic equations for the macroscopic variables and thus to close the system of electrohydrodynamics for nematic elastomers we start from the balance equations for all macroscopic variables. For the conserved quantities density, density of linear momentum and concentration we have

$$\dot{\rho} + \nabla_i g_i = 0 \tag{4.1}$$

$$\dot{g}_i + \nabla_j \sigma_{ij} = 0 \tag{4.2}$$

$$\dot{c} + \nabla_i j_i^c = 0 \tag{4.3}$$

where σ_{ij} is the stress tensor and j_i^c the concentration current. For the nonconserved fields the balance equations take the form

$$\dot{\epsilon}_{ij} + X_{ij} = 0 \tag{4.4}$$

$$\dot{n}_i + Y_i = 0 \tag{4.5}$$

$$\dot{S} + Z = 0 \tag{4.6}$$

$$\dot{\tilde{\Omega}}_i + Y_i^\Omega = 0 \tag{4.7}$$

$$\dot{D}_i + J_i^P = 0 \tag{4.8}$$

$$\dot{\sigma} + \nabla_i j_i^\sigma = \frac{R}{T} \tag{4.9}$$

In eqs.(4.4) - (4.9) X_{ij} and Y_i are the quasi-currents of the variables associated with broken translational (network) and rotational (director) symmetry. To guarantee rotational invariance of the dynamic equation for the strain field, one must require $X_{ij} = X_{ji}$. Z and Y_i^Ω are the quasi-currents linked to temporal changes of the order parameter and of

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relative rotations, respectively. J_i^P is the polarization current and j_i^σ the entropy current. The second law of thermodynamics requires positivity of the source term in eq.(4.9), i.e. of the entropy production R/T, for dissipative processes. For reversible effects the dissipation function R must vanish identically.

Of eq. (4.8) we will use the divergence part only, $\dot{\nabla}_i D_i + \nabla_i J_i^P = 0$, which is the charge conservation law, while we will discard the two complementary variables $curl\,\mathbf{D}$, which are relevant only for temporally rapidly varying electromagnetic fields. There is no a priori reason why these very fast variables should be slowed down to hydrodynamic time scales in elastomers. For the same reason we do not consider dynamical equations for the magnetic fields. Within this assumption $curl\,\mathbf{E}$ will be eliminated in the entropy production (4.19) and only symmetric gradients of $\mathbf{E}$ occur.

To close the system of macroscopic dynamic equations, one must link the currents and quasi-currents in eqs.(4.1)-(4.9) to the thermodynamic forces discussed in the last section. To achieve this we split all currents and quasi-currents into reversible ($R = 0$) and irreversible ($R > 0$) contributions and discuss the two types separately.

We obtain for the linear, reversible parts of the currents, making use of general symmetry and invariance arguments

$$g_i^R = \rho_0 v_i \tag{4.10}$$

$$\sigma_{ij}^R = p\,\delta_{ij} - \psi_{ij} + \beta_{ij}W - \frac{\lambda^\perp}{2}(L_j^\perp n_i + L_i^\perp n_j) - \frac{1}{2}(1-\lambda)n_i h_j$$
$$+ \frac{1}{2}(1+\lambda)n_j h_i \tag{4.11}$$

$$j_i^{cR} = c_0 v_i \tag{4.12}$$

$$X_{ij}^R = -A_{ij} \tag{4.13}$$

$$Y_i^R = -\epsilon_{ijk}\omega_j n_k - \lambda(\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j)A_{jk} \tag{4.14}$$

$$Z^R = \beta_{ij}A_{ij} \tag{4.15}$$

$$Y_i^{\Omega R} = -\frac{\lambda^\perp}{2}(\delta_{ij}^\perp n_k + \delta_{ik}^\perp n_j)A_{jk} \tag{4.16}$$

$$J_i^{PR} = \rho_{el} v_i \tag{4.17}$$

$$j_i^{\sigma R} = \sigma_0 v_i \tag{4.18}$$

with the abbreviations $A_{ij} = \frac{1}{2}(\nabla_i v_j + \nabla_j v_i)$ and $\omega_i = \frac{1}{2}\epsilon_{ijk}\nabla_j v_k$. The reversible cross-coupling β_{ij} between the modulus and velocity gradients is of the uniaxial symmetric form $\beta_{ij} = \beta_\parallel n_i n_j + \beta_\perp \delta_{ij}^\perp$.

To derive the dissipative contributions to the currents it is most convenient to start with the expression for the dissipation function R. The dissipative currents are then obtained by taking variational derivatives with respect to one thermodynamic conjugate while keeping all others fixed. Expanding the dissipation function R to terms up to second order in the thermodynamic forces we obtain

$$
\begin{aligned}
R = R_0 + \int d\tau \Big[& \frac{1}{2}\gamma_{ij}(\nabla_k \psi_{ik})(\nabla_l \psi_{jl}) + \frac{1}{2}\kappa_w W^2 + \frac{1}{2\gamma_1}(\delta_{ij}^\perp h_i h_j)^2 \\
& + (\nabla_j \psi_{ij})(\zeta_{ik}^T \nabla_k T + \zeta_{ik}^c \nabla_k \mu_c + \zeta_{ik}^W \nabla_k W + \zeta_{ik}^E E_k) + \zeta_{ijk}^E E_k \nabla_j h_i \\
& + \zeta_{12}\delta_{ij}^\perp L_i^\perp h_j + \frac{1}{2}\zeta^\perp L_i^\perp L_j^\perp \delta_{ij}^\perp + \frac{1}{2}\sigma_{ij}^E E_i E_j + \kappa_{ij}^E E_i(\nabla_j T) + D_{ij}^E E_i(\nabla_j \mu_c) \\
& + \lambda_{ij}^{WT}(\nabla_j W)(\nabla_i T) + \lambda_{ij}^{W\mu}(\nabla_j W)(\nabla_i \mu_c) + \zeta_{ijk}^{\Omega E}(\nabla_i E_j)L_k^\perp \Big]
\end{aligned}
\tag{4.19}
$$

where R_0 is the part of the dissipation function already present in binary fluid mixtures of miscible liquids.

In eq.(4.19) all second rank tensors, i.e. γ_{ij}, ζ_{ij}^T, ζ_{ij}^c, ζ_{ij}^W, ζ_{ij}^E, κ_{ij}^E, D_{ij}^E, λ_{ij}^T and $\lambda_{ij}^{W\mu}$ are of the uniaxial form $\alpha_{ij} = \alpha_\parallel n_i n_j + \alpha_\perp \delta_{ij}^\perp$. In addition to strain diffusion they include coupling terms between strain gradients and gradients of temperature, chemical potential of mixtures and of the order parameter molecular field and to electric fields as well. The molecular field associated with relative rotations, $L_i^\perp$, couples to the molecular field of the director ($\propto \zeta_{12}$), to gradients of the electric field ($\propto \zeta_{ijk}^{\Omega E}$) and relaxes ($\propto \zeta^\perp$). The third rank tensors in eq.(4.19), $\zeta_{ijk}^{\Omega E}$, ζ_{ijk}^E are of the form (assuming $curl\,\mathbf{E} = 0$)

$$\zeta_{ijk} = \zeta(\delta_{ik}^\perp n_j + \delta_{jk}^\perp n_i) \tag{4.20}$$

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and thus bring along only one dissipative coefficient each.

The dissipative parts of the currents are then obtained by taking the variational derivative of the dissipation function with respect to the appropriate thermodynamic force while keeping all other forces fixed (indicated by ...). We find for example

$$Z^D = \frac{\delta R}{\delta W}\big|_{...} = \kappa_w W - \zeta_{ik}^W \nabla_k \nabla_j \psi_{ij} - \lambda_{ij}^{WT} \nabla_j \nabla_i T - \lambda_{ij}^{W\mu} \nabla_j \nabla_i \mu_c \tag{4.21}$$

$$Y_i^D = \frac{-\delta R}{\delta \nabla_j \Phi_{ij}}\big|_{...} = +\frac{1}{\gamma_1} h_j \delta_{ij}^\perp - \zeta_{12} L_i^\perp \tag{4.22}$$

$$X_{ij}^D = \frac{\delta R}{\delta \psi_{ij}}\big|_{...} = \tag{4.23}$$

$$-\frac{1}{2}\Big[\nabla_j\big(\gamma_{ik}\nabla_l\psi_{kl} + \zeta_{ik}^T\nabla_k T + \zeta_{ik}^c\nabla_k\mu_c + \zeta_{ik}^W\nabla_k W + \zeta_{ik}^E E_k\big) + (i \leftrightarrow j)\Big]$$

$$Y_i^{\Omega D} = \frac{\delta R}{\delta L_i^\perp}\big|_{...} = \zeta^\perp L_i^\perp + \zeta_{12} h_i + \zeta_{kji}^{\Omega E}\nabla_k E_j \tag{4.24}$$

$$J_i^{PD} = \frac{\delta R}{\delta E_i}\big|_{...} = \sigma_{ij}^E E_j + \kappa_{ij}^E(\nabla_j T) + D_{ij}^E(\nabla_j \mu_c) - \zeta_{jik}^{\Omega E}\nabla_j L_k^\perp \tag{4.25}$$

5. Conclusions and Perspective

In this article we have given the hydrodynamic and electrohydrodynamic equations for nematic side-chain elastomers. We have emphasized the importance of relative rotations on both, the static and the dynamic properties of nematic elastomers. The generalization of the work presented here to other elastomeric liquid crystalline phases is under way and will be presented elsewhere.

In contrast to nematic side-chain polymers the displacement field **u** turns out to be a true hydrodynamic variable in nematic elastomers due to the presence of the permanent network. Nevertheless there can be free ends of polymer chains in permanently cross-linked networks, which might be connected dynamically to form an additional transient network absent in the static limit and for sufficiently small frequencies. These processes will give rise to additional dynamic degrees of freedom, which become relevant at frequencies above the characteristic frequency for the dynamic glass transition. They will again have to be

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described by the addition of the strain tensor associated with this transient network to the list of macroscopic variables just as for ordinary polymers [42] and for nematic side-chain polymers [43,44]. Thus the present description does not give as a special case a polymer melt.

Another important issue is the question of the spatial homogeneity on smaller length scales. Recent static and dynamic experiments in the groups of Finkelmann and Strobl show, that the modulus and orientation of the nematic order parameter in the immediate vicinity of the cross-linking points can influence substantially the macroscopic properties of nematic elastomers. In the present paper we have assumed that the nematic order in the ground state of the elastomer is spatially homgeneous. Thus our approach corresponds to a coarse-grained description of these materials. What seems to be highly desirable on the long term is a mesoscopic description forming a bridge between the macroscopic approach presented here and the various microscopic pictures, e.g. from simulations, which are not able as yet to accomodate the relevant timescales necessary for the description of macroscopic phenomena as discussed in this paper.

Note added

Since this work has been finished, conventional rubber elasticity has been generalized to nematic elastomers [54] to incorporate cubic coupling terms involving the electric field, director gradients and the Cauchy strain tensor. These cubic coupling terms correspond to a strain dependence of the flexoelectric terms, that is to one possible nonlinear generalization of the first term in the fourth line of eq.(3.2).

Acknowledgements

Appendix

In the bulk of this manuscript we have used the electric displacement vector $\mathbf{D}$ as the macroscopic variable and the (internal) electric field $\mathbf{E}$ then follows as a thermodynamic conjugate by derivation of the energy. However, in the presence of external fields it is more convenient to take $\mathbf{E}$ as variable and $\mathbf{D}$ as conjugate. This is acchieved by switching from the energy density ϵ to the electric enthalpy density $\tilde{\epsilon}$ via a Legendre transformation $\tilde{\epsilon} = \epsilon - \mathbf{E} \cdot \mathbf{D}$ [52]. The Gibbs relation then reads

$$T d\sigma = d\tilde{\epsilon} - \mu d\rho - \mu_c dc - v_i dg_i - W dS + h_i d\,n_i - \psi_{ij} d\epsilon_{ij} - L_i^\perp d\tilde{\Omega}_i + D_i dE_i \qquad (A.1)$$

and the electric enthalpy density is written as

$$\begin{aligned}
\tilde{\epsilon} = &\frac{1}{2} c_{ijkl} \epsilon_{ij} \epsilon_{kl} + (\chi_{ij}^\rho \delta\rho + \chi_{ij}^\sigma \delta\sigma + \chi_{ij}^c \delta c + \chi_{ij}^S \delta S) \epsilon_{ij} \\
&+ \frac{1}{2} \tilde{K}_{ijkl} (\nabla_j n_i)(\nabla_l n_k) + \frac{a}{2} (\delta S)^2 + (b^\sigma \delta\sigma + b^\rho \delta\rho + b^c \delta c) \delta S \\
&- \frac{1}{2} \epsilon_{ij}^E E_i E_j + \frac{1}{2} \chi_{ij} H_i H_j + \frac{1}{2} D_1 \tilde{\Omega}_i \tilde{\Omega}_i + D_2 \tilde{\Omega}_i \epsilon_{jk} n_j \delta_{ik}^\perp \\
&+ \tilde{e}_{ijk} (\nabla_i n_j) E_k + \tilde{e}_{ij}^S (\nabla_i \delta S) E_j + \tilde{M}_{ijk} (\nabla_i n_j)(\nabla_k \delta S) \\
&+ \tilde{\chi}_{ijkl}^E E_i E_j \epsilon_{kl} + \chi_{ijkl}^H H_i H_j \epsilon_{kl} + \tilde{\chi}_{ijkl}^N (\nabla_i E_j) \epsilon_{kl} \\
&+ \tilde{\gamma}_{ijk}^E E_i E_j \tilde{\Omega}_k + \gamma_{ijk}^H H_i H_j \tilde{\Omega}_k + \tilde{\gamma}_{ijk}^N \tilde{\Omega}_k \nabla_i E_j
\end{aligned} \qquad (A.2)$$

The material tensors denoted by a tilde in eq.(A.2) have the same form as their counterparts (without the tilde) in eq.(3.2), but the susceptibility coefficients (with the tilde) contained in the tensors are taken at constant $\mathbf{E}$ and are generally different from those in sec.3 (for the coefficients which are equal to those of sec.3 within the given expansion we have omitted the tilde in eq.(A.2)). We have included χ_{ijkl}^E and χ_{ijkl}^H, the electrostrictive and magnetostrictive tensors familiar from classical solids [50], which comprise six electro-(magneto-) strictive coefficients in a uniaxial material (compare, for example, refs. [22] and [50] for the explicit structure of the tensor). For nematic elastomers there are electro- and magneto-strictive effects with respect to relative rotations ($\propto \gamma_{ijk}^{E,H}$) in addition to those more familiar ones with respect to elastic strains.

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Invoking $curl\,\mathbf{E} = 0$ as a static Maxwell condition the tensor $\tilde{\chi}_{ijkl}^N$ is symmetric with respect to the first two indices and, thus, of the same structure as the electrostrictive tensor, while $\tilde{\gamma}_{ijk}^N = \tilde{\gamma}_{jik}^N$ implies $\tilde{\gamma}_{ijk}^N = \tilde{\gamma}(\delta_{jk}^{\perp}n_i + \delta_{ik}^{\perp}n_j)$. Performing in $\tilde{e}_{ijk}E_k\nabla_i n_j$ a partial integration, one obtains the same expression up to a global divergence. Therefore one never has to deal, for the flexoelectric terms, with gradients of $\mathbf{E}$, and thus, there is no $curl\,\mathbf{E}$ contribution in the flexo-electric terms. However, if one *linearizes* in the director and its gradients, then $\tilde{e}_1 = \tilde{e}_3$ in the bulk follows from partial integration and symmetrization of $\nabla_j E_i$ [53].

The generalized forces are obtained from eq.(A.2) by variational derivation, e.g. $D_i \equiv -(\delta/\delta E_i)\int \tilde{\epsilon}\,d\tau$ while keeping all other variables constant

$$L_i^{\perp} = D_1\tilde{\Omega}_i + D_2\epsilon_{jk}n_j\delta_{ik}^{\perp} + \tilde{\gamma}_{kji}^E E_j E_k + \gamma_{kji}^H H_j H_k + \tilde{\gamma}_{kji}^N \nabla_k E_j \qquad (A.3)$$

$$\psi_{ij} = c_{ijkl}\epsilon_{kl} + \chi_{ij}^{\rho}\delta\rho + \chi_{ij}^{\sigma}\delta\sigma + \chi_{ij}^{c}\delta c + \chi_{ij}^{S}\delta S + \frac{1}{2}D_2(\tilde{\Omega}_i n_j + \tilde{\Omega}_j n_i)$$
$$+ \tilde{\chi}_{klij}^E E_k E_l + \chi_{klij}^H H_k H_l + \tilde{\chi}_{klij}^N \nabla_k E_l \qquad (A.4)$$

$$h_i = \delta_{ip}^{\perp}\nabla_j\left(\tilde{K}_{pjkl}\nabla_l n_k + \tilde{e}_{jpk}E_k + \tilde{M}_{jpk}\nabla_k\delta S\right)$$
$$- \frac{1}{2}\delta_{iq}^{\perp}\left(\epsilon_{pj}\epsilon_{kl}\frac{\partial c_{pjkl}}{\partial n_q} + (\nabla_j n_p)(\nabla_l n_k)\frac{\partial K_{pjkl}}{\partial n_q} + 2D_2\left(\tilde{\Omega}_q\epsilon_{jk}n_j n_k - \tilde{\Omega}_j\epsilon_{qk}\delta_{jk}^{\perp}\right)\right)$$
$$- \left(2\delta_{ij}^{\perp}n_p n_k - \delta_{pj}^{\perp}\delta_{ik}^{\perp} - \delta_{jk}^{\perp}\delta_{ip}^{\perp}\right)\left(\tilde{M}(\nabla_p n_j)(\nabla_k S) + \tilde{\gamma}\,\tilde{\Omega}_j\nabla_p E_k\right)$$
$$- n_j\delta_{ip}^{\perp}\left(\epsilon_a E_p E_j - \chi_a H_p H_j\right)$$
$$+ \left(\delta_{ik}^{\perp}n_j + \delta_{ij}^{\perp}n_k\right)\left(\left[\chi_a^{\rho}\delta\rho + \chi_a^c\delta c + \chi_a^S\delta S\right]\epsilon_{kj} + \tilde{e}_a^S E_j\nabla_k\delta S\right) \qquad (A.5)$$

$$W = a\delta S + \chi_{ij}^S\epsilon_{ij} + b^{\sigma}\delta\sigma + b^{\rho}\delta\rho + b^c\delta c - \nabla_k(\tilde{M}_{ijk}\nabla_i n_j) - \nabla_j(\tilde{e}_{ij}^S E_i) \qquad (A.6)$$

$$D_i = \epsilon_{ij}^E E_j - \tilde{e}_{kji}\nabla_k n_j - \tilde{e}_{ij}^S\nabla_j S - 2\tilde{\chi}_{ijkl}^E E_j\epsilon_{kl} + \tilde{\chi}_{jikl}^N \nabla_j\epsilon_{kl}$$
$$- 2\tilde{\gamma}_{ijk}^E E_j\tilde{\Omega}_k + \nabla_j(\tilde{\gamma}_{jik}^N \tilde{\Omega}_k) \qquad (A.7)$$

where in eq.(A.5) only the relevant transverse part of h_i is considered. For the remaining thermodynamic conjugates μ, μ_c and v_i one can proceed in the same way.

Except for the additional strictive terms, the eqs.(A.3)-(A.7) are completely equivalent to eqs.(3.4)-(3.8), where $\mathbf{D}$ has been chosen as variable. A direct comparison of these

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two sets of equations gives the relations between the static susceptibilities at constant $\mathbf{E}$ (with tilde) to those at constant $\mathbf{D}$ (without tilde).

$$\tilde{e}_{\parallel}^S = \epsilon_{\parallel}^E e_{\parallel}^S \qquad\qquad\qquad \tilde{e}_{\perp}^S = \epsilon_{\perp}^E e_{\perp}^S$$

$$\tilde{e}_1 = \epsilon_{\parallel}^E e_1 \qquad\qquad\qquad \tilde{e}_3 = \epsilon_{\perp}^E e_3$$

$$\tilde{M} = M - e_1 e_{\parallel}^S \epsilon_{\parallel}^E = M - e_3 e_{\perp}^S \epsilon_{\perp}^E \qquad\qquad \tilde{K}_2 = K_2 \qquad\qquad (A.8)$$

$$\tilde{K}_1 = K_1 - \epsilon_{\parallel}^E e_1^2 \qquad\qquad\qquad \tilde{K}_3 = K_3 - \epsilon_{\perp}^E e_3^2$$

$$\tilde{\gamma} = \epsilon_{\parallel}^E \gamma_1 = \epsilon_{\perp}^E \gamma_2 \qquad\qquad\qquad \tilde{\chi}_{ijkl}^N = \epsilon_{jp}^E \chi_{ipkl}^N$$

If one would require at this point $\tilde{e}_1 = \tilde{e}_3$, this would imply the unphysical result $e_{\parallel}^S = e_{\perp}^S$

In the true thermodynamic equilibrium all the thermodynamic conjugates on the left hand sides of eqs.(A.3)-(A.7) (or equivalently eqs.(3.4)- (3.8)) have to vanish, which immediately gives $\delta S = E_i = \tilde{\Omega}_i = \epsilon_{ij} = 0$ with constant, but arbitrary director orientation (suppressing density, concentration and thermal degree of freedom for the moment). Applying an external constant electric field (and assuming that the local electric field $\mathbf{E}$ is also constant in this situation) the stationary state is given by $L_i^{\perp} = \psi_{ij} = h_i = W = 0$ and e.g. $E_i = E_0 \delta_{iz}$. The solution of these equations shows that the director $\mathbf{n}$ is either parallel or perpendicular to $\mathbf{E}$ (depending on the sign of ϵ_a), that there is a finite elongational strain ϵ_{ij}^0, a change in the nematic order parameter δS^0, but zero relativ rotation $\tilde{\Omega}_i^0 = 0$. For the case $\mathbf{E} \parallel \mathbf{n}$ we find

$$\epsilon_{zz}^0 = E_0^2 N_{\parallel} R^{-1}$$

$$\epsilon_{\perp\perp}^0 = E_0^2 N_{\perp} R^{-1}$$

$$\delta S^0 = -a^{-1} E_0^2 (\chi_{\parallel}^S N_{\parallel} + \chi_{\perp}^S N_{\perp}) R^{-1} \qquad\qquad (A.9)$$

$$D_z^0 = \epsilon_{\parallel}^E E_0 + O(E_0^3)$$

where

$$N_{\parallel} = \tilde{\chi}_{42}^E (c_4 - a^{-1} \chi_{\parallel}^S \chi_{\perp}^S) - \tilde{\chi}_3^E (c_1 - a^{-1} \chi_{\perp}^{S\,2})$$

$$N_{\perp} = \tilde{\chi}_3^E (c_4 - a^{-1} \chi_{\parallel}^S \chi_{\perp}^S) - \tilde{\chi}_{42}^E (c_3 - a^{-1} \chi_{\parallel}^{S\,2}) \qquad\qquad (A.10)$$

$$R = (c_1 - a^{-1} \chi_{\perp}^{S\,2})(c_3 - a^{-1} \chi_{\parallel}^{S\,2})(c_4 - a^{-1} \chi_{\parallel}^S \chi_{\perp}^S)^2$$

A quite similar situation arises in an external magnetic field. To elastomers not only electric or magnetic external fields can be applied in order to orient the director, also externally imposed strains are an effective tool for that purpose. In that case an equilibrium state requires $L_i^\perp = h_i = W = D_i = 0$. Inspecting eq. (A.5) one finds that the torque on $\mathbf{n}$ vanishes, if for a purely elongational external strain the director is parallel or perpendicular to the dilation axis (depending on the sign of some linear combination of elastic constants that are connected with the elastic anisotropy energy). In this case also a change in the order parameter is induced. For an external shear strain $\mathbf{n}$ is perpendicular to the shear plane and no change of the order parameter is obtained. For elastomers linear elasticity is known to be a poor description even for small strains. The addition of cubic contributions $(\propto c_{ijklmp}^{(3)} \epsilon_{ij} \epsilon_{kl} \epsilon_{mp})$ turns out, however, to give good agreement with experimental results [28] performed in the isotropic phase above a nematic-isotropic transition in a number of nematic elastomers. This observation, together with the appropriate crosscouplings to the other variables, opens the possibility that different orientations of the director are energetically favoured depending on the strength of the external strain. Details will be given elsewhere.

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