

## Nonlinear effects in the electrohydrodynamics of uniaxial nematic liquid crystals

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We study the influence of nonlinear effects on the electrohydrodynamics of uniaxial nematic liquid crystals. To measure a novel reversible nonlinear coupling between an electric field, distortions of the director, and the stress tensor, we suggest an experimental setup and predict the occurrence of a flow in an initially static configuration. We show that the dissipative effects proposed by R. S. Akopyan and B. Ya. Zeldovich [Sov. Phys.—JETP **60**, 953 (1985)] do not exist.

### I. INTRODUCTION

The hydrodynamic description of uniaxial nematic liquid crystals is by now well established<sup>1-6</sup> and has been checked experimentally in detail.<sup>3,7,8</sup> The study of electrohydrodynamic effects has been triggered by the application of electro-optic effects in these materials in display devices.<sup>9</sup> Nevertheless it seems clear that the combination of hydrodynamics and electric effects<sup>10</sup> has still not been achieved to a satisfactory extent on the nonlinear level.

In the present paper we derive nonlinear electrohydrodynamic equations for uniaxial nematics, thus generalizing the nonlinear hydrodynamic equations presented previously.<sup>3-6</sup> Static as well as dynamic phenomena will be considered and an experiment to test a novel nonlinear cross coupling between gradients of the velocity field, the macroscopic polarization, and gradients of the director will be discussed.

When incorporating electric effects, one must keep in mind that hydrodynamic excitations can acquire a gap for small wave vectors due to the long-range nature of the Coulomb forces, which render Goldstone's theorem inapplicable.

Naturally the incorporation of the macroscopic polarization is not only important for the low-molecular-weight, rodlike, thermotropic nematic phases predominantly studied so far, but will be at least as important for the investigation of the physical properties of lyotropic nematic phases and nematic liquid crystalline polymers, the study of which is just starting.

Having in mind the applicability of the equations derived to lyotropic phases, we also keep as a macroscopic variable, i.e., as a quantity which relaxes in a long, but finite, time  $\tau$ , the modulus of the nematic order parameter  $S$ . These variations can be expected to be crucial in thermotropic nematics mainly close to the isotropic transition and in the vicinity of defects, but for lyotropic materials they are probably much more important since one can expect that external forces such as electric and magnetic fields or shear flow will lead to a local change in the concentration of the micelles (which are embedded in an iso-

tropic background fluid) and thus to a local change of the degree of order. Therefore we think that the influence of the spatial variations of the modulus on the macroscopic behavior cannot be overestimated for lyotropic systems. As we go along we will also consider at each step the influence of mixture effects, i.e., we incorporate an additional concentration as conserved quantity. Such effects are obviously always present in lyotropic nematics as those are always multicomponent systems (typically one has water, a higher alcohol, and a surfactant as constituents).

The present paper is organized as follows. In Sec. II we discuss static properties, in Sec. III we investigate reversible and dissipative dynamic effects followed by the suggestion of an experimental setup for the study of a nonlinear reversible current in Sec. IV. In Sec. V we present the discussion and the conclusions including an analysis of why the dissipative terms suggested by Akopyan and Zeldovich<sup>11</sup> do not exist. The latter is based on a time-reversal symmetry argument. In the Appendix we summarize new static and dynamic contributions to the electrohydrodynamics of other liquid crystalline phases such as cholesterics, hexatic-*B*, smectic-*I* and -*F*, chiral smectic phases, etc.

### II. STATICS

To derive the general electrohydrodynamic equations, one has to determine first the macroscopic variables. Those come in two groups: truly hydrodynamic variables and macroscopic quantities which relax in a long, but finite, time  $\tau$ . For uniaxial nematics we have the hydrodynamic variables density  $\rho$ , entropy density  $\sigma$ , density of linear momentum  $\mathbf{g}$ , and concentration of one component (in binary mixtures) along with the variations  $\delta \mathbf{n}$  of the director  $\hat{\mathbf{n}}$  characterizing the spontaneously broken continuous rotational symmetry of uniaxial nematics. As additional macroscopic variables we have the macroscopic displacement  $\mathbf{D}$  (or, equivalently, its thermodynamic conjugate, the local electric field  $\mathbf{E}$ ) and the modulus

of the order parameter  $S$  which is introduced via  $Q_{ij} = S(n_i n_j - \frac{1}{3} \delta_{ij})$  with  $n_i^2 = 1$ . In thermotropic nematics, the modulus is especially important as a macroscopic variable close to the nematic-isotropic transition and in the vicinity of defects, where the modulus varies as a function of space. The Gibbs relation takes the form

$$T d\sigma = d\varepsilon - \mu d\rho - \mathbf{V} \cdot d\mathbf{g} + \mathbf{D} \cdot d\mathbf{E} - Z dc - W dS - h_i dn_i, \quad (1)$$

with  $\mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}$ , where  $\mathbf{E}$  is the internal field, which will be, in general, different from the external field.

For the generalized energy we find

$$\begin{aligned} \Phi = \Phi_0 + \Phi_1 + \int d\tau \{ & [\lambda^\rho(\partial_i \rho) + \lambda^\sigma(\partial_i \sigma) + \lambda^c(\partial_i c)] [\delta_{ik}^{\text{tr}} n_j + \delta_{jk}^{\text{tr}} n_i] (\partial_j n_k) + L_{\parallel} (n_i \partial_i S)^2 + L_{\perp} \delta_{ij}^{\text{tr}} (\partial_i S) (\partial_j S) \\ & + M (\partial_i S) (\partial_j n_k) (\delta_{ik}^{\text{tr}} n_j + \delta_{jk}^{\text{tr}} n_i) + (\zeta_1 \delta_{ik}^{\text{tr}} n_j + \zeta_2 \delta_{jk}^{\text{tr}} n_i) E_i \partial_j n_k + (\zeta_3 \delta_{ij}^{\text{tr}} + \zeta_4 n_i n_j) E_i \partial_j S \\ & + H_{ij}^{\rho} (\partial_j \rho) E_i + H_{ij}^{\sigma} (\partial_j \sigma) E_i + H_{ij}^c (\partial_j c) E_i \} , \end{aligned} \quad (2)$$

where the terms-coupling gradients of the director on one hand and gradients of the density, the concentration, the entropy density, and the modulus on the other lead in addition to surface terms of the structure

$$\int d\sigma_i n_i n_j \partial_j \rho, \quad \int d\sigma_i n_i n_j \partial_j c,$$

etc. In Eq. (1)  $\Phi_0$  contains all the standard terms, i.e., those already given, e.g., in Ref. 3, including the Frank free energy and the dielectric term, whereas  $\Phi_1$  contains all the contributions which are not related to electric effects, distortions of  $\hat{\mathbf{n}}$ , and which are not contained in Ref. 3:

$$\Phi_1 = \int d\tau [\beta_1 (\delta S)^2 + \beta_2 (\delta S) (\delta \rho) + \beta_3 (\delta S) (\delta c) + \beta_4 (\delta S) (\delta \sigma) + N_{ij}^{\rho} (\partial_i S) (\partial_j \rho) + N_{ij}^{\sigma} (\partial_i S) (\partial_j \sigma) + N_{ij}^c (\partial_i S) (\partial_j c)] . \quad (2a)$$

The  $\lambda^i$  terms have been introduced as higher-order gradient terms in Ref. 5 for  $\rho$  and  $\sigma$ ; here we have also incorporated the effect of an additional concentration. The gradient energy for the modulus ( $\sim L_{\parallel}, L_{\perp}$ ) takes the standard form for an uniaxial system. The cross-coupling term  $M$  between  $\nabla S$  and  $\nabla \mathbf{n}$  has been discussed first from a general point of view in Ref. 12. For a discussion in the framework of a Ginzburg-Landau free energy, cf. Ref. 13; it is clear, however, that in general there is no simple relationship between the values of the coefficients (e.g., between  $L_{\parallel}$ ,  $L_{\perp}$  and  $M$ ). These simple relations are artifacts of the Ginzburg-Landau approximation.  $\zeta_1$  and  $\zeta_2$  represent the usual flexoelectric contributions and  $\zeta_3$  and  $\zeta_4$  come from a term  $E_i \partial_j Q_{kl}$  which has been given for the first time in Ref. 14 and which gives  $\zeta_1$ ,  $\zeta_2$ ,  $\zeta_3$ , and  $\zeta_4$  when specialized to uniaxial nematic liquid crystals. As already noted in Ref. 14, the  $\zeta$  terms contain both gradients of the director and gradients of the modulus. The last two lines written down in Eq. (2) represent higher-order gradient terms coupling the electric field to gradients of all scalar conserved densities, i.e., to density, entropy density, and concentration. These contributions have not been given before.

By inspection of Eq. (2) we see immediately that every macroscopic variable except for the density of linear momentum (which has a different behavior under time reversal) is coupled statically to every other variable if higher-order gradient terms are taken into account.

We would like to stress that the generalized energy does not contain any nontrivial terms to cubic order in the variables, where we mean by trivial terms those which contain only the scalar variables [as, e.g.,  $(\delta \rho)^2 \delta \sigma$ ] or which are straightforward generalizations of quadratic terms such as, e.g.,

$$\varepsilon_{ij} E_i E_j \rightarrow (\varepsilon_{ij} + \varepsilon_{ij}^{\rho} \delta \rho + \varepsilon_{ij}^{\sigma} \delta \sigma + \varepsilon_{ij}^c \delta c + \varepsilon_{ij}^S \delta S) E_i E_j . \quad (3)$$

The thermodynamic forces are obtained from the generalized energy written down in Eq. (2) by taking the variational derivative with respect to one variable while keeping all the other ones fixed, e.g.,

$$h_i = \delta \Phi / \delta n_i |_{E, \rho, \dots} . \quad (4)$$

### III. DYNAMICS

To derive the dynamic couplings we relate the currents and quasicurrents which appear in the balance equations for the macroscopic variables to the thermodynamic forces which in turn have been obtained in Sec. II by taking variational derivatives of the generalized energy with respect to the variables.

For the balance equations we have

$$\begin{aligned} \dot{\rho} + \partial_i g_i &= 0, \\ \dot{g}_i + \partial_j \sigma_{ij} &= 0, \\ \dot{n}_i + Y_i &= 0, \\ \dot{S} + X &= 0, \\ \dot{P}_i + j'_i &= 0, \\ \dot{\sigma} + \partial_i j_i^{\sigma} &= \frac{R}{T}, \end{aligned} \quad (5)$$

where  $R$  is the entropy production. In Eq. (5) we have written down a dynamic equation for the macroscopic polarization which has to be supplemented by Maxwell's

equations along the lines discussed in Ref. 15. The conservation law for the energy density follows from the other balance equations in (5), because all hydrodynamic variables are linked by the Gibbs relation (1).

For the dissipative terms it is most convenient to write down the entropy production  $R$  and then to determine the dissipative parts of the currents and quasicurrents by taking variational derivatives of  $R$  with respect to one ther-

modynamic force while keeping all the other quantities at a fixed value, e.g.,

$$\sigma_{ij}^D = \frac{\delta R}{\delta \nabla_i v_j} \Big|_{\dots}$$

We find

$$\begin{aligned} R = R_0 + \int d\tau [ & \nu W^2 + \mu_{ij}^{(1)} (\nabla_i W) (\nabla_j T) + \mu_{ij}^{(2)} (\nabla_i W) E_j + \sigma_{ij}^E E_i E_j \\ & + \sigma'_{ij} E_i (\nabla_j T) + (\zeta_{ijk}^{(2)} (\nabla_j T) + \zeta_{ijk}^{(3)} E_j) h_i E_k + \psi_{mqij}^{(2)} (\nabla_i n_j) E_q h_m + \gamma_{ijk}^{(1)} h_k \nabla_i E_j \\ & + \gamma_{ijk}^{(2)} (\nabla_i \nabla_j T) h_k + \tau_{ijkl}^{(2)} W^2 (\nabla_i n_j) (\nabla_k n_l) + \tau_{ijklmn}^{(3)} (\nabla_i n_j) (\nabla_k n_l) (\nabla_m T) E_n + \tau_{ijklmn}^{(4)} (\nabla_i n_j) (\nabla_k n_l) E_m E_n ] , \end{aligned} \quad (6)$$

where  $R_0$  is the entropy production discussed before<sup>6</sup> without electric fields and spatial variations of the modulus  $S$ . For the structure of  $\zeta_{ijk}^{(2)}$  and  $\zeta_{ijk}^{(3)}$  we have<sup>16</sup>

$$\begin{aligned} \zeta_{ijk}^{(2)} &= \zeta_1^{(2)} \delta_{ik}^{\text{tr}} n_j + \zeta_2^{(2)} \delta_{ij}^{\text{tr}} n_k , \\ \zeta_{ijk}^{(3)} &= \zeta^{(3)} (\delta_{ik}^{\text{tr}} n_j + \delta_{ij}^{\text{tr}} n_k) \end{aligned} \quad (7)$$

and where  $\mu_{ij}^{(1)}$  and  $\mu_{ij}^{(2)}$  have uniaxial form. The latter terms are associated with linear dissipation and mean that spatial variations of the modulus are dissipatively coupled to electric and/or temperature gradients, thus opening additional channels for dissipation. For the higher-order gradient terms  $\gamma_{ijk}^{(i)}$  we have

$$\begin{aligned} \gamma_{ijk}^{(1)} &= \gamma_1^{(1)} \delta_{ik}^{\text{tr}} n_j + \gamma_2^{(1)} \delta_{jk}^{\text{tr}} n_i , \\ \gamma_{ijk}^{(2)} &= \gamma^{(2)} (\delta_{ik}^{\text{tr}} n_j + \delta_{ij}^{\text{tr}} n_k) . \end{aligned} \quad (8)$$

Aside from the nonlinear dissipative terms written down explicitly in Eq. (6), all linear coefficients can be made to be nonlinear in a straightforward way by keeping explicitly a linear dependence on the modulus, e.g.,  $\mu_{ij} = \mu_{ij}^{(0)} + \mu_{ij}^{(1)} W$ . To incorporate the effects of an additional concentration all one has to do is to replace  $\nabla_i T$  by  $\nabla_i Z$  and to keep the Soret effect.

We note that the the cross-coupling term  $M$  between gradients of the director and gradients of the modulus (in the generalized energy) has no counterpart in the dissipation function; a term  $\sim h_i W$  does not exist. For the reversible currents we find

$$\begin{aligned} \alpha_{ijklp} &= \alpha_1 (n_i \delta_{jk}^{\text{tr}} \delta_{lp}^{\text{tr}} + n_j \delta_{ik}^{\text{tr}} \delta_{lp}^{\text{tr}}) + \alpha_2 n_k \delta_{ij}^{\text{tr}} \delta_{lp}^{\text{tr}} + \alpha_3 (n_i \delta_{ji}^{\text{tr}} \delta_{kp}^{\text{tr}} + n_j \delta_{ii}^{\text{tr}} \delta_{kp}^{\text{tr}}) + \alpha_4 n_l \delta_{ij}^{\text{tr}} \delta_{kp}^{\text{tr}} \\ &+ \alpha_5 (n_i \delta_{jp}^{\text{tr}} \delta_{kl}^{\text{tr}} + n_j \delta_{ip}^{\text{tr}} \delta_{kl}^{\text{tr}}) + \alpha_6 n_i n_j n_l \delta_{kp}^{\text{tr}} + \alpha_7 (n_k \delta_{ij}^{\text{tr}} \delta_{pj}^{\text{tr}} + n_k \delta_{ip}^{\text{tr}} \delta_{jl}^{\text{tr}}) + \alpha_8 n_i n_j n_k \delta_{pl}^{\text{tr}} \\ &+ \alpha_9 (\delta_{ik}^{\text{tr}} \delta_{jp}^{\text{tr}} + \delta_{ip}^{\text{tr}} \delta_{jk}^{\text{tr}}) n_l + \alpha_{10} n_k n_l (\delta_{ip}^{\text{tr}} n_j + \delta_{jp}^{\text{tr}} n_i) , \end{aligned} \quad (10)$$

i.e., there are ten independent phenomenological parameters contained in  $\alpha_{ijklp}$ , if we require the stress tensor to be symmetric.<sup>19</sup> Thus if one starts with a deformed director configuration, all macroscopic variables are dynamically coupled. It seems important to note that upon linearization the new terms ( $\sim \alpha_{ijklp}$ ) vanish identically.

$$\begin{aligned} j_k^{\sigma R} &= -\alpha_{ijklp}^{(\sigma)} (\nabla_i V_j) (\nabla_l n_p) , \\ j_k^{E R} &= -\alpha_{ijklp}^{(E)} (\nabla_i V_j) (\nabla_l n_p) , \\ X^R &= \beta_{ij} \nabla_i V_j , \\ Y^R &= \frac{1}{2} (1 - \lambda) \delta_{ij}^{\text{tr}} n_k - (1 + \lambda) \delta_{ik}^{\text{tr}} n_j , \\ \sigma_{ij}^R &= \beta_{ij} W + \lambda (n_i h_j + n_j h_i) + \frac{1}{2} (n_i h_j - n_j h_i) \\ &+ \phi_{kj} \nabla_i n_k + \frac{\varepsilon}{4\pi} \left[ E_i E_j - \frac{E^2}{2} \delta_{ij} \right] \\ &+ \alpha_{ijklp}^{(\sigma)} (\nabla_k T) (\nabla_l n_p) + \alpha_{ijklp}^{(E)} E_k (\nabla_l n_p) , \end{aligned} \quad (9)$$

where the linear terms ( $\sim \lambda, \frac{1}{2}$ ) are the ones familiar from the hydrodynamics of uniaxial nematics. In addition one has contributions  $\sim \beta_{ij}$  coupling gradients of the velocity field to the modulus—these are isomorphic to terms considered before for a coupling to the smectic or discotic order parameter close to the nematic-smectic  $A$  (Ref. 17) or nematic-columnar<sup>18</sup> transitions. The novel terms are  $\sim \alpha_{ijklp}^{(E)}$  and  $\alpha_{ijklp}^{(\sigma)}$ . They represent nonlinear cross couplings between velocity gradients, gradients of the director, and electric fields or temperature gradients, i.e., if an electric field is applied to a sample with a distorted director field, the first equation in (9) predicts the appearance of flow. This will be discussed in more detail in Sec. IV.

The same effect does not only arise for temperature gradients or electric fields but also for concentration gradients in mixtures. We find for the structure of  $\alpha_{ijklp}^{(i)}$ :

This situation is different from that for the flow alignment term  $\lambda$  coupling the director to extensional flow, which survives when linearized. The latter coupling, which was the first reversible current bringing along a phenomenological parameter, namely,  $\lambda$ , has been studied in great detail for uniaxial nematics, both theoretical-

ly<sup>20,1,2</sup> and experimentally.<sup>21-23</sup> More recently<sup>24,25</sup> flow alignment has also been investigated for the coupling to the in-plane director in smectic *C*. This record of the investigation of reversible dynamic effects gives rise to the expectation that purely nonlinear effects can also be observed.

#### IV. PROPOSED EXPERIMENT

To investigate experimentally the nonlinear reversible cross coupling discussed in Sec. IV, it would be very important to have a simple experimental setup for a critical check of their magnitude. To be specific we consider a "hybrid" cell,<sup>26</sup> i.e., a layer of nematics between two electrodes with the alignment of the director being planar on one electrode and homeotropic on the other. In addition we will focus on the case of a temporally constant electric field. All considerations below are equally valid for an externally applied temperature or concentration gradient.

From Eq. (9) we have

$$\sigma_{ij}^R(\alpha^{(E)}) = \alpha_{ijklp}^{(E)} E_k (\nabla_l n_p) \quad (11)$$

or for  $\dot{g}_i$

$$\begin{aligned} \dot{g}_i(\alpha^{(E)}) &= -\nabla_j \sigma_{ij} \\ &= -\nabla_j (\alpha_{ijklp}^{(E)} E_k \nabla_l n_p). \end{aligned} \quad (12)$$

By inspection of Eq. (12) we see immediately that the hybrid configuration chosen is very convenient to detect  $\dot{g}_i$  because it is clear from Eq. (12) that even for a spatially constant external electric field, finite second spatial derivatives of the director field will be sufficient to produce flow phenomena. And a hybrid cell provides us by construction with finite first, second, and higher-order derivatives of the director in a static configuration (as long as no external field is applied).

To demonstrate the effect to be expected in more detail, we assume for simplicity the global configuration to be two-dimensional with the normal to the plates along the *z* direction and the director lying in the *z-x* plane. In addition we suppose that the sample has a sufficiently large extent in *y* direction, so that the spatial variation in this direction can be neglected at least initially. Then it is clear that there is no twist and everything happens in the *x-z* plane. We find from Eq. (11) that this also produces flow only in the *x-z* plane. Nevertheless all coefficients contribute to the resulting flow pattern and it seems very difficult to suggest a setup which shows the influence of one of the coefficients only. At this stage, however, it is most important to establish the effect predicted qualitatively. If the field is applied in a direction which has also a *y* component, however, a fully three-dimensional flow is to be expected in the hybrid cell due to the new cross couplings.

#### V. DISCUSSION AND CONCLUSIONS

In this paper we have analyzed the nonlinear electrohydrodynamics of uniaxial nematic liquid crystals and we have found a number of new dynamic couplings, both dissipative and reversible. To detect a novel nonlinear reversible cross coupling between extensional flow, external

field, and gradients of the director field, we have suggested a simple experimental setup.

We would like to point out here that the nonlinear "dissipative" terms given in a recent paper by Akopyan and Zeldovich<sup>11</sup> do not exist. To see this, all one has to do is to study the behavior of Eq. (A4) in Ref. 11 under time reversal. As is well known (cf., e.g., Ref. 2), the dissipation function *R* is invariant under time reversal. Inspecting Eq. (A4) of Ref. 11, one sees immediately that all contributions are products of three terms. And in all contributions two of the three terms are invariant under time reversal whereas the third one is always odd. Thus all contributions written down on the right-hand side are odd under time reversal and can therefore not be dissipative. We mention for completeness that none of the 12 contributions written down in Eq. (A4) of Ref. 11 leads to vanishing entropy production and would therefore be a candidate for a reversible term. Only the terms presented in this paper are nonlinear reversible cross-coupling terms between extensional flow, gradients of the director field, and external temperature gradients.

In the Appendix we have written down a number of new static and dynamic contributions for the electrohydrodynamics of other liquid crystalline phases. Especially we have discussed the analogs of the new terms for uniaxial nematics for the bond angle in hexatic-*B* and smectic-*I* and -*F* phases.

In complementing the derivation of the macroscopic dynamics presented here, one of us<sup>27</sup> has given very recently the dynamics close to the nematic and cholesteric to isotropic transitions in an external electric field. There we find again a similar type of nonlinear reversible cross-coupling term as the one presented here. Amusingly enough it turns out that close to the cholesteric to isotropic transition it is even possible to have a coupling which has one gradient less ( $\alpha \sim \underline{Q}E$  with  $\underline{Q}$  the order parameter of the transition), because one has a pseudoscalar quantity available ( $q_0$ ).

In deriving the macroscopic dynamics presented here we have kept as a macroscopic variable the modulus of the orientational order parameter. As mentioned earlier this will be mainly important for lyotropic multicomponent systems where spatial inhomogeneity effects for the modulus can be expected to play an important role under the influence of external forces such as electric fields, temperature and concentration gradients, shear, etc.

We note that the effects described can only be observed for dc and low-frequency ac fields. If the frequency is too high, not all variables can follow. Finally we stress that none of the effects linear in the electric field has a magnetic counterpart due to the different behavior of magnetic and electric fields under parity and time reversal.

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## APPENDIX

In this appendix we summarize new results for the electrohydrodynamics of various liquid crystalline phases such as cholesterics, chiral smectic, hexatic-*B* (cf. Ref. 28 for the hydrodynamics), etc. For the liquid crystalline phases with bond-orientational order, i.e., for hexatic-*B* and for smectic-*I* and -*F*, we find that there are no flexoelectric terms due to the bond angle. One has only the standard flexoelectric terms associated with the layering which is isomorphic to that of the *A* phase<sup>29</sup> for hexatic *B* and to that of the *C* phase<sup>15</sup> for smectic *I* and *F*. For the chiral counterparts of smectic *I* and *F*, i.e., for *I*<sup>\*</sup> and *F*<sup>\*</sup>, flexoelectric effects are isomorphic to those given for *C*<sup>\*</sup>.

In closing the discussion on static effects we mention that terms of the form

$$\int d\tau \{ [\zeta_i^p \delta\rho + \zeta_i^\sigma \delta\sigma + \zeta_i^c \delta c + \zeta_i^s \delta S + \zeta_i^u (\nabla_{||} u_{||})] E_i \} \quad (\text{A1})$$

with  $\zeta_i^k = \zeta^k q_0 \hat{p}_i$  exist for smectic *I*<sup>\*</sup> and *F*<sup>\*</sup>. They are thus of the same structure as for the piezoelectric effect in cholesterics and smectic *C*<sup>\*</sup> (Ref. 15) and as the couplings

$$R^{(3)} = R_{E=0}^{(3)} + \int d\tau [\kappa_{ijk}^{(2)} (\nabla_i T) (\nabla_j T) E_k + \kappa_{ijk}^{(3)} (\nabla_i T) E_j E_k + \kappa_{ijk}^{(4)} E_i E_j E_k + \tilde{\eta}_{ijklm} (\nabla_i V_k) (\nabla_j V_l) E_m + \tilde{\tau}_{ijk}^{(1)} (\nabla_i T) E_k (\nabla_j \phi) + \tilde{\tau}_{ijk}^{(2)} E_i E_j (\nabla_k \phi) + \tilde{\tau}_{ijk}^{(3)} E_i (\nabla_j \phi) (\nabla_k \phi)], \quad (\text{A4})$$

which is a generalization of Eq. (2.13) of Ref. 10. From inspection of Eq. (A4) it is evident that in the dissipation function every macroscopic variable in cholesterics couples to every other one in cubic order. It is straightforward to incorporate the effect of an impurity concentration in Eq. (A4). All one has to do is to replace  $\nabla_i T$  by  $\nabla_i Z$  and to add the couplings of concentration and temperature. The situation in cholesterics is very different from that in smectic *A* where we do not find any additional electrohydrodynamic terms in cubic order. For chiral smectic phases such as *C*<sup>\*</sup>, *I*<sup>\*</sup>, and *F*<sup>\*</sup>, terms isomorphic to those written down for cholesterics exist.

For the reversible currents we find in hexatic *B* for the analog of the  $\alpha_{ijklp}$  in uniaxial nematics

$$\sigma_{ij}^{(\alpha)} = \alpha_{ijkl} (\nabla_k T) (\nabla_l \Theta) \quad (\text{A5})$$

with

to the scalar conserved quantities introduced very recently.<sup>14</sup>

For the electrohydrodynamic contributions to the dissipation function, the analysis presented in the main part of the present paper can be easily extended to cover other liquid crystalline phases. Specifically we obtain for hexatic *B*

$$R = \int d\tau \{ \nu_{ijkl} A_{ij} A_{kl} + \kappa_{ij} (\nabla_i T) (\nabla_j T) + \mu (\nabla_i \chi_i)^2 + \xi (\nabla_i \phi_i)^2 + \xi \hat{p}_i (\nabla_i T) (\nabla_i \phi_i) + \beta \chi_i \hat{p}_i (\nabla_i \phi_i) (\nabla_m \chi_m) + [\beta_{iq} (\nabla_q T) + \tilde{\beta}_{iq} E_q] \chi_i (\nabla_i \chi_i) \}. \quad (\text{A2})$$

In Eq. (A2) all second-rank tensors are of the form

$$\gamma_{iq} = \gamma_{||} \hat{p}_i \hat{p}_q + \gamma_{\perp} \delta_{iq}^{\text{tr}}. \quad (\text{A3})$$

We have also incorporated in Eq. (A2) nonlinear terms that have not been previously considered and which contain derivatives of the bond angle.

For cholesterics we obtain

$$\alpha_{ijkl} = \alpha (\epsilon_{ikl} p_j + \epsilon_{jkl} p_i), \quad (\text{A6})$$

i.e., instead of ten coefficients for uniaxial nematics we find only one for the bond angle in hexatic *B*.<sup>30</sup> For smectic *I* and *F* we find that  $\alpha_{ijkl}$  contains ten independent parameters for the bond angle  $\Theta$  and ten parameters for  $\varphi$ .

For cholesterics and for all chiral smectic phases (*C*<sup>\*</sup>, *F*<sup>\*</sup>, and *I*<sup>\*</sup>) we have for the nontrivial contributions to the stress tensor

$$\sigma_{ij}^R = \zeta_{ijk}^{(1)} E_k q_0 + \zeta_{ijk}^{(2)} \nabla_k T q_0, \quad (\text{A7})$$

where<sup>31</sup>

$$\zeta_{ijk}^{(l)} = \zeta_1^{(l)} p_i p_j p_k + \zeta_2^{(l)} (p_j \delta_{ik}^{\text{tr}} + p_i \delta_{jk}^{\text{tr}} + p_k \delta_{ij}^{\text{tr}}). \quad (\text{A8})$$

An analogous term exists also for concentration gradients. If the requirement of  $\sigma_{ij}$  being symmetric is dropped, we find that  $\zeta_{ijk}^{(l)}$  has six independent components.

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