

Maxwell equations in nematic liquid crystals

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A complete set of hydrodynamic equations, rigorously describing the behavior of both nematic liquid crystals and electromagnetic fields, is derived and presented. The field dissipation in nematics is rather more complicated than could be accounted for by ϵ and μ being complex. The symmetric, total stress tensor includes field anisotropy, flexoelectricity, and the ponderomotive force. Boundary conditions and experimental consequences are outlined.

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The macroscopic Maxwell equations [1]

$$\dot{\mathbf{D}} = c\nabla \times \mathbf{H}^M - \mathbf{j}^e, \quad \dot{\mathbf{B}} = -c\nabla \times \mathbf{E}^M \quad (1)$$

can be conveniently considered as the equations of motion for \mathbf{D} and \mathbf{B} , two fields that are constrained to satisfy $\nabla \cdot \mathbf{D} = \rho^e$, $\nabla \cdot \mathbf{B} = 0$ at all times. The fields \mathbf{H}^M and \mathbf{E}^M then need to be expressed in \mathbf{D} and \mathbf{B} to render Eqs. (1) closed. Usually, one either takes \mathbf{H}^M and \mathbf{E}^M as real functions of \mathbf{B} and \mathbf{D} , which neglects dissipation in insulators, or takes $\mathbf{H}^M = \mathbf{B}/(\mu_R + i\mu_I)$, $\mathbf{E}^M = \mathbf{D}/(\epsilon_R + i\epsilon_I)$ for a given frequency, valid only in the linear regime of weak fields. Recently, the nonlinear yet dissipative relationships

$$\mathbf{H}^M = \mathbf{H} - \alpha c\nabla \times \mathbf{E}, \quad \mathbf{E}^M = \mathbf{E} + \beta c\nabla \times \mathbf{H} \quad (2)$$

were derived for the stationary, isotropic, polarizable and magnetizable liquid [2]. Note the difference between \mathbf{H}^M , \mathbf{E}^M , and $\mathbf{H} \equiv \partial\epsilon/\partial\mathbf{B}$, $\mathbf{E} \equiv \partial\epsilon/\partial\mathbf{D}$, respectively, ϵ being the energy density: The former are the general, nonequilibrium (Maxwell) fields, defined by Eqs. (1), the latter are equations of state and contain only equilibrium information. Generally, \mathbf{H} and \mathbf{E} are nonlinear functions of all the thermodynamic variables, especially \mathbf{D} and \mathbf{B} . The dissipative terms $\alpha c\nabla \times \mathbf{E}$ and $\beta c\nabla \times \mathbf{H}$ are of different parity under time reversal than \mathbf{H} and \mathbf{E} , respectively, and account for damping in Eqs. (1) irrespective of the constitutive equations. In the linear regime, Eqs. (2) naturally reduce to a complex ϵ and μ : Expanding \mathbf{H} and \mathbf{E} for an electromagnetic wave of amplitude $D \sim B$ and of frequency $\omega = cq$, one obtains $\mu_R = B/H$, $\epsilon_R = D/E$, $\mu_I = \omega\alpha\mu_R^2$, $\epsilon_I = \omega\beta\epsilon_R^2$.

There is another, maybe more important advantage of Eqs. (2): it can be readily generalized to systems with broken symmetries. Denoting

$$\mathbf{H}^M = \mathbf{H} + \mathbf{H}^D, \quad \mathbf{E}^M = \mathbf{E} + \mathbf{E}^D, \quad (3)$$

with \mathbf{D} referring to dissipation, \mathbf{E}^D and \mathbf{H}^D are generally more complicated than in Eqs. (2) and depend on additional dissipative forces. In nematics, we shall find the gradient of the temperature ∇T , the shear flow v_{ij} , and the nematic molecular field Ψ to contribute linearly to \mathbf{E}^D and \mathbf{H}^D . Obviously, this fact cannot be accounted for by the quantities μ_I and ϵ_I , however weak the fields are. [This more general dependence may be considered

as the nonequilibrium extension of flexoelectricity [3–5], which accounts for the thermodynamic dependence of \mathbf{E} on $\nabla_i n_j$, cf. Eq. (6) below.]

The experimental consequences of these couplings are numerous and warrant detailed study. Foremost is the possibility to create or modify external fields by inhomogeneities in temperature, velocity and Ψ ; or vice versa, to alter the latter by applying electric or magnetic fields. Then there are ramifications for stationary situations, in which suitable boundary conditions induce finite ∇T , v_{ij} or Ψ . Being proportional to these, \mathbf{H}^D , $\mathbf{E}^D \neq \mathbf{0}$ even in the absence of applied fields. As a result, the static Maxwell equations $\nabla \times \mathbf{H} = \mathbf{0}$, $\nabla \times \mathbf{E} = \mathbf{0}$ are not valid, yet are customarily employed [6–8].

The derivation, presentation, and consideration of the Maxwell equations in the nematics, more specifically of the expressions for \mathbf{H}^D and \mathbf{E}^D , is the purpose of this paper. However, this task can, and will, only be tackled simultaneously with the study of nematodynamics in the presence of fields. Especially, the total stress tensor (including field anisotropy, flexoelectricity, the Maxwell tensor and its dissipative part) is derived and shown to be symmetric. As a corollary, a hydrodynamically exact expression for the ponderomotive force, valid even out of equilibrium [3,8,9], is obtained. Part of it is, rather naturally, proportional to the spatial derivative of \mathbf{E}^D and \mathbf{H}^D . The paper is divided into four sections: The introductory part ends here. Next is statics, including thermodynamics, equilibrium conditions, and the identification of the irreversible thermodynamic forces, both for the fields and the nematic degrees of freedom. Dynamics is third, with separate considerations for the reactive and dissipative terms in the hydrodynamic field equations, including the Maxwell equations. The expressions for \mathbf{E}^D , \mathbf{H}^D , and for the total stress tensor are to be found here. Finally, experimental consequences and boundary conditions are briefly discussed.

The statics. The energy density ϵ is a function of the director \mathbf{n} , its spatial derivative, the fields \mathbf{B} and \mathbf{D} , and of the densities of mass ρ , entropy s , and momentum \mathbf{g}

$$d\epsilon = \mu d\rho + T ds + \mathbf{v} \cdot d\mathbf{g} + \psi_{ij} d\nabla_j n_i + \varphi_i dn_i + \mathbf{H} \cdot d\mathbf{B} + \mathbf{E} \cdot d\mathbf{D}. \quad (4)$$

(Note that \mathbf{g} is not the conserved, total momentum density, $\mathbf{g}^{\text{tot}} = \mathbf{g} + \mathbf{D} \times \mathbf{B}/c = \rho\mathbf{v} + \mathbf{E} \times \mathbf{H}/c$. The difference

results from the Lorentz transformation behavior of the fields and was discussed in detail in Ref. [9]: \mathbf{H} and \mathbf{E} are the respective conjugate variables of B and D only if \mathbf{g} is held constant; holding instead \mathbf{g}^{tot} constant, the conjugate variables would be the respective fields in the rest frame.) In equilibrium, maximizing the total entropy with appropriate constraints, we obtain a constant chemical potential μ and the vanishing of all the thermodynamic forces: ∇T , $\mathbf{n} \times \Psi$, $\nabla \times \mathbf{H}^0$, $\nabla \times \mathbf{E}^0$, v_{ij} [2,4,5]. The following notations apply: the molecular field $\Psi_i \equiv \varphi_i - \nabla_j \psi_{ij}$, the shear flow $v_{ij} = (\nabla_i v_j + \nabla_j v_i)/2$, and the superscript 0 denotes rest frame quantities. Off equilibrium, the rate R of entropy production is a quadratic function of these forces. In conductors, $\mathbf{E}^0 = \mathbf{0}$ supplants $\nabla \times \mathbf{E}^0 = \mathbf{0}$; in bad conductors both terms are needed: the latter is more quickly reached,

while the first accounts for the true equilibrium condition.

Rotating the coordinate system, or equivalently all the vectors and tensors of Eq. (4), must not change the energy density. This yields the useful rotation identity [4], valid even out of equilibrium:

$$(\mathbf{E} \times \mathbf{D} + \mathbf{H} \times \mathbf{B} + \mathbf{v} \times \mathbf{g} + \varphi \times \mathbf{n})_i + \varepsilon_{ijk}(\psi_{lj} \nabla_k n_l + \psi_{jl} \nabla_l n_k) = 0. \quad (5)$$

Assuming weak fields, we can obtain concrete expressions for the conjugate variables Ψ , \mathbf{H} , \mathbf{E} , \dots by expanding the rest frame energy $\Delta\varepsilon = \varepsilon^0 - \varepsilon^0(\rho, s)$ to second order in $\nabla_i n_j$, \mathbf{D}^0 , and \mathbf{B}^0 ,

$$\Delta\varepsilon^0 = [K_{ijkl} \nabla_i n_j \nabla_k n_l + (\mu^{-1})_{ij} B_i^0 B_j^0 + (\varepsilon^{-1})_{ij} D_i^0 D_j^0]/2 + \varepsilon_1^f n_i D_i \nabla_j n_j + \varepsilon_2^f D_i n_j \nabla_j n_i \quad (6)$$

and then taking the respective derivative. [The susceptibilities K_{ijkl} , $(\mu^{-1})_{ij}$, and $(\varepsilon^{-1})_{ij}$ have the usual form [4,5].] The hydrodynamic equations given below, however, are valid independent of Eq. (6). The flexoelectric coefficients ε_i^f enter the constitutive equation $\mathbf{E}(\mathbf{D})$ and allow for an \mathbf{E} field even if \mathbf{D} vanishes [4,5,10]. The flexoelectric coefficients (along with the field anisotropy) also enter the molecular field Ψ , rendering it dependent on \mathbf{D} . This fact (the neglect of which conveys the wrong impression that the stress tensor is not symmetric) does not seem to be widely recognized [8]. Flexomagnetism enters Eq. (6) in higher than the considered order.

The Dynamics. Given the thermodynamics, Eq. (4), and the irreversible forces, the hydrodynamic field equations can be derived employing the ‘‘standard hydrodynamic procedures’’: calculate $\dot{\varepsilon}$ via Eq. (4) and require the result to become $\nabla \cdot \mathbf{Q}$, where \mathbf{Q} is the energy flux. This is indeed how the usual nematodynamics [4,5,11] and the Maxwell equations for isotropic liquids [2] were derived. A simultaneous derivation is what has been carried out here. The resultant equations, in addition to the Maxwell equations, Eqs. (1) and (3), are

$$\dot{\rho} + \nabla(\rho\mathbf{v}) = 0, \quad (7)$$

$$\mathbf{n} \times [\dot{\mathbf{n}} + (\mathbf{v}\nabla)\mathbf{n} + \mathbf{n} \times (\nabla \times \mathbf{v})/2] + \mathbf{Y}^D = \mathbf{0},$$

$$\dot{s} + \nabla(s\mathbf{v} - \mathbf{f}^D) = R/T,$$

$$\partial_t(\mathbf{g} + \mathbf{D} \times \mathbf{B}/c)_i + \nabla_j(\Pi_{ij} - \Pi_{ij}^D) = 0, \quad (8)$$

where the reactive part of the stress tensor (of the total and conserved momentum density $\mathbf{g}^{\text{tot}} = \mathbf{g} + \mathbf{D} \times \mathbf{B}/c$) is given as

$$\Pi_{ij} = (Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g} + \mathbf{E} \cdot \mathbf{D} + \mathbf{H} \cdot \mathbf{B} - \varepsilon)\delta_{ij} + g_i v_j - E_i D_j - H_i B_j + \Pi_{ij}^{\text{nem}}, \quad (9)$$

and as usual, $\Pi_{ij}^{\text{nem}} = \nabla_i n_k \psi_{kj} + (\Psi_j n_i - \Psi_i n_j)/2$. To show that Π_{ij} can be brought into an equivalent symmetric form, $\Pi_{ij} = \Pi_{ji}$, one can rewrite Π_{ij}^{nem} as

$$\nabla_i n_k \psi_{kj} + \{\psi_{jk} \nabla_k n_i - \psi_{ik} \nabla_k n_j + n_i \varphi_j - n_j \varphi_i - \nabla_k[(n_i \psi_{kj} - n_k \psi_{ij}) + (i \leftrightarrow j)]\}/2$$

of which the very last term ($i \leftrightarrow j$) was added in by hand, permissible since $\nabla_j \nabla_k (i \leftrightarrow j) = 0$. With Eq. (5), Π_{ij} now takes the symmetrized form

$$(Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g} + \mathbf{E} \cdot \mathbf{D} + \mathbf{H} \cdot \mathbf{B} - \varepsilon)\delta_{ij} + \{g_i v_j - E_i D_j - H_i B_j + \nabla_i n_k \psi_{kj} - \nabla_k [n_i \psi_{kj} - n_k \psi_{ij}]\}/2 + \{i \leftrightarrow j\}.$$

Equations (1,3,8) can be combined to obtain the proper expression for the ponderomotive force

$$\rho d_t(g_i/\rho) + \nabla_i(Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g} - \varepsilon) + E_j \nabla_i D_j + H_j \nabla_i B_j = (\rho^e \mathbf{E} + j^e \times \mathbf{B}/c)_i - \nabla_j(\Pi_{ij}^{\text{nem}} - \Pi_{ij}^D) + (\mathbf{B} \times \nabla \times \mathbf{H}^D + \mathbf{D} \times \nabla \times \mathbf{E}^D)_i. \quad (10)$$

The first term on the left, with the material derivative $d_i = \partial_i + v_i \nabla_i$, contains both the acceleration and the Abraham force [3]. The second term appears to be the simple generalization of the pressure, while the third and fourth act, respectively, as part of the electric and magnetic ponderomotive force. This *interpretation*, however, is far from unique: These three terms combine to yield

$$\begin{aligned} \nabla_i(Ts + \mu\rho + \mathbf{v} \cdot \mathbf{g} - \epsilon) + E_j \nabla_i D_j + H_j \nabla_i B_j \\ = s \nabla_i T + \rho \nabla_i \mu + g_j \nabla_i v_j - \psi_{kj} \nabla_i n_k - \varphi_j \nabla_i n_j, \end{aligned}$$

another innocuously looking generalization of (the gradient of) the pressure. (The formula, of course, remains unique and is not adversely affected by the ambiguity in identifying the pressure, not really a useful concept in the presence of fields.) New are the last two terms on the right of Eq. (10), containing H^D and E^D , respectively, which may be interpreted as the dissipative part of the ponderomotive force; (cf. Eqs. (12) below for the explicit forms of H^D and E^D). The counter part in condensed matter of the Lorentz force is obviously given by the first term, so the force a charged object experiences is $\sim E$ rather than E^M [12].

The explicit form of the dissipative terms in Eqs. (1,3,7,8), all denoted with superscript D , are obtained from the rate R of entropy production,

$$\begin{aligned} R = \mathbf{f}^D \cdot \nabla T + \Pi_{ij}^D v_{ij} + \mathbf{Y}^D \cdot (\mathbf{n} \times \Psi) + \mathbf{j}^D \cdot \mathbf{E}^0 \\ + \mathbf{E}^D \cdot (c \nabla \times \mathbf{H}^0) - \mathbf{H}^D \cdot (c \nabla \times \mathbf{E}^0) \end{aligned} \quad (11)$$

of which the fluxes are expanded in the forces. (The expression for R is also a result of the hydrodynamic standard procedure; $\mathbf{j}^D = \mathbf{j}^e - \rho^e \mathbf{v}$ is the dissipative electric current in the rest frame.) In the limit of vanishing equilibrium fields, where only the anisotropy of the nematic director needs to be considered, a first order expansion yields

$$\begin{pmatrix} E_i^D \\ f_i^D \\ j_i^D \end{pmatrix} = \begin{pmatrix} \beta_{ij} & \xi_{ij} & 0 \\ -\xi_{ji} & \kappa_{ij} & \gamma_{ij} \\ 0 & \gamma_{ji} & \sigma_{ij} \end{pmatrix} \begin{pmatrix} (c \nabla \times \mathbf{H}^0)_j \\ \nabla_j T \\ E_j^0 \end{pmatrix}, \quad (12)$$

$$\begin{pmatrix} -H_i^D \\ Y_i^D \\ \Pi_{ik}^D \end{pmatrix} = \begin{pmatrix} \alpha_{ij} & \eta \delta_{ij}^T & \lambda_{ijl} \\ \eta \delta_{ji}^T & \delta_{ij}^T / \gamma_1 & \zeta_{ijl} \\ -\lambda_{jik} & -\zeta_{jik} & \nu_{ikjl} \end{pmatrix} \begin{pmatrix} (c \nabla \times \mathbf{E}^0)_j \\ (\mathbf{n} \times \Psi)_j \\ v_{jl} \end{pmatrix}, \quad (13)$$

where $\delta_{ij}^T \equiv \delta_{ij} - n_i n_j$, $\lambda_{ijl} = \lambda(\epsilon_{ikj} n_k n_l + [j \leftrightarrow l])$, and $\zeta_{ijl} = (\gamma_2 / 2\gamma_1)(\epsilon_{ikj} n_k n_l + [j \leftrightarrow l])$, ν_{ikjl} have the usual forms [4,5,11]. All second rank tensors remain of the structure $\alpha_{ij} = \alpha_\perp \delta_{ij}^T + \alpha_\parallel n_i n_j$. Of special interests are η , λ , ξ_\perp , and ξ_\parallel of which only η is dissipative. They couple thermodynamic forces to the Maxwell equations, and vice versa, the field dissipation to nematodynamics.

In the presence of finite equilibrium fields, arbitrarily oriented with respect to \mathbf{n} , the system is biaxial and possesses many more independent elements in the force-flux relation. In the absence of ordering and clarifying experimental data, or of model calculations, the exercise

of classification, therefore, becomes somewhat pointless. However, one should note that a finite equilibrium E field allows a coupling that invalidates the above block diagonal form,

$$E_i^D = \mu_{ijl} v_{jl} + \dots, \quad \text{where} \quad (14)$$

$$\begin{aligned} \mu_{ijl} = \mu_1 E_i \delta_{jl}^T + \mu_2 E_i n_j n_l + \mu_3 (E_j \delta_{il}^T + E_l \delta_{ij}^T) \\ + \mu_4 (E_j n_i n_l + E_l n_i n_j) + \mu_5 E_i E_j E_l. \end{aligned} \quad (15)$$

A finite H field is not as consequential.

Finally, we turn our attention to the *experimental consequences* and outline the qualitative features here. First, we examine the boundary conditions: Given Eqs. (1), it is clear that (barring singular surface charges or currents) the usual condition of continuity holds for the normal components of \mathbf{D} , \mathbf{B} and for the parallel ones of \mathbf{E}^M , \mathbf{H}^M . A direct consequence of this is: If the nematic surface is parallel to \mathbf{E}^M , \mathbf{H}^M , it is possible to produce fields outside the nematic liquid even if \mathbf{E} , \mathbf{H} vanish within; alternatively, one may modify any pre-existing fields. For instance, to produce or modify an E field outside, one may either enforce a temperature gradient or a shear flow. As Eqs. (12) and (14) indicate, these two effects are parameterized, respectively, by the transport matrices ξ_{ij} and μ_{ijl} . Flow alignment is easily seen as contributing to μ_{ijl} : A static E field sustains a nonvanishing polarization, say along the director \mathbf{n} . A shear flow perpendicular to \mathbf{n} with a gradient along the field will rotate the polarization somewhat out of the equilibrium orientation and induce a perpendicular component, which in turn produces the desired perpendicular electric field E_\perp outside the nematics. Assuming a simple relaxation equation for the polarization \mathbf{P} ,

$$\dot{\mathbf{P}} + \mathbf{P} \times \Omega = \delta \mathbf{P} / \tau,$$

where Ω is the vorticity and $\delta \mathbf{P}$ the deviation from equilibrium, one finds $\epsilon_0 E_\perp = P_\parallel \tau \nabla_\parallel v_\perp / 2$, with P_\parallel the equilibrium polarization. Given a large enough value of the relaxation time τ , the perpendicular field E_\perp should be easily measurable. If the flow alignment is the only mechanism contributing to the transport coefficient μ_3 , we may identify it with $P_\parallel \tau / 2\epsilon_0$, which reduces to $E\tau(\epsilon_0 - \epsilon) / 2\epsilon_0$ for a linear constitutive relation.

This effect vanishes if the static E field does, but the analogous H -field effect, given by λ_{ijl} of Eq. (13), does not. One mechanism for the zero-field effect could be of steric origin, as in flexoelectricity; another would arise from the alignment of the director's permanent magnetic moment by the vorticity of the shear flow. The magnitude of the first is difficult to evaluate, while the second is probably much smaller than the finite-field effect, although detection is possibly facilitated by the absence of any equilibrium fields.

If, on the other hand, the nematic surface is perpendicular to D or B , then the field outside is strictly given by the one inside, independent of any thermodynamic forces such as temperature gradients or shear flows. So interestingly, in the hydrodynamic theory outlined here, the variables D and B pair up with the more complicated

E^M and H^M , containing both thermodynamic and dissipative information, to yield the respective vacuum fields.

Clearly, it is physically and experimentally relevant to distinguish E from E^M , and H from H^M . Two more examples: The Ohmic current is driven by $\mathbf{E}^0 = \mathbf{E} + \mathbf{v} \times \mathbf{B}/c$, cf. Eqs. (12), so is the Lorentz force of, and discussed below, Eq. (10). On the other hand, if $\dot{B} = 0$, we have $\nabla \times \mathbf{E}^M = \nabla \times (\mathbf{E} + \mathbf{E}^D) = \mathbf{0}$ and there is always a potential Φ^M , with $\mathbf{E}^M = -\nabla\Phi^M$. (However, if $\nabla \times \mathbf{E}^D$ does not vanish, this potential must not be taken as proportional to the Coulomb potential $-\int d^3x \rho^e / 4\pi r$.)

In more general situations, one has to solve the hydrodynamic equations for a given set of boundary conditions — for all hydrodynamic variables including the electromagnetic field. Even if these boundary conditions are all stationary, oscillatory instabilities occasionally set in if the deviation from equilibrium exceeds a threshold [6,7]. As a result, the static Maxwell equations no longer hold yet are usually employed. (The rationale presumably stems from the constitutive equation $D = \epsilon(T, n_i)E$. So even if T or n_i are time dependent, the effect on D

or E is nonlinearly small. However, as should have become clear by now, this constitutive equation is incorrect, since D and B participate fully in the linear hydrodynamics.) What is more, even in the basic, unstructured stationary state, before any instability sets in, the clear ramification of Eqs. (12–14) is that despite stationarity $\nabla \times \mathbf{E} \neq \mathbf{0}$, $\nabla \times \mathbf{H} \neq \mathbf{j}^e$; rather, one must include $\nabla \times \mathbf{E}^D$ and $\nabla \times \mathbf{H}^D$.

Two concluding remarks: (i) The approach chosen here to study the electromagnetic field in nematics is rather general, based solely on the concepts of irreversible thermodynamics and broken symmetries. Therefore, the qualitative features of most results pertain to other anisotropic systems as well, such as smectics, cholesterics, crystals, or the two phases of superfluid ^3He . The specific symmetry of a given system will of course permit different thermodynamic and transport tensors. (ii) The Maxwell equations presented here are only valid in the hydrodynamic regime, confined to $\omega\tau_i \ll 1$, where τ_i includes any microscopic relaxation time, especially that of the polarization and magnetization. This clearly excludes relevance to optical frequency phenomena.

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- [1] The Heaviside-Lorentz system of units employed in this paper is by far the most convenient one in the present context. It is essentially the Gaussian system, but with all four fields reduced, and all four sources increased, by the factor of $2\sqrt{\pi}$. In other words, $B^H = B^G/2\sqrt{\pi}$, similarly for D, H, E ; while $\rho^H = 2\sqrt{\pi}\rho^G$, similarly for j, P, M .
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- [12] The discussion in this paragraph pertains to any continuous media, irrespective of broken symmetries. The difference to the formulas of Ref. [9], which predates Ref. [2], arise from the fact that the two terms E^D and H^D were conventionally and erroneously set to zero.