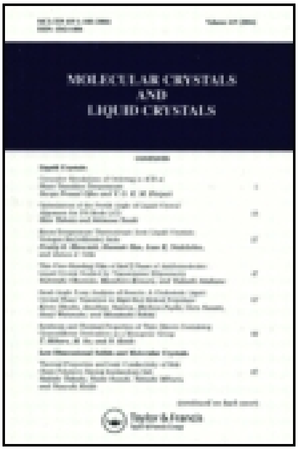

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Some Nonlinear Electrohydrodynamic Effects in Nematic Liquid Crystals

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The nonlinear extension of the standard electrohydrodynamic equations for a single mode, in the nematic phase, results in a set of coupled nonlinear equations coupling the space charge density and the local curvature. As a result of the nonlinear form, it is possible to predict, ab-initio, the existence of periodicity. When one uses assumptions of a simplifying nature on the field quantities, the resulting formalism is capable of encompassing, in addition to the usual voltage threshold predictions for the linear approximation, equations which are similar in form to Landau's equation for velocity amplitudes of turbulent fluids suggesting the possibility of a direct explanation of the dynamic scattering mode. Other ramifications of the nonlinear theory of electrohydrodynamics are also discussed.

The problem of the response of nematic liquid crystals to externally applied electric and magnetic fields has received a great deal of attention in recent years.[1–7] Most of the theories, to date, have used formulations which ignore the intrinsic nonlinear character of the basic equations, and in the process, may have lost the capacity to describe a host of possible effects. Notable exceptions include the work of Carroll,[5] Ben-Abraham,[6] and of Pikin, *et al.*[7] Carroll[5] presents initial discussions of nonlinear effects which include the dependence of alignment angles on voltages above threshold, while Ben-Abraham[6] sets up a nonlinear sine-Gordon equation for the torque balance equation in terms of the angle θ (between the director and the x-axis). Pikin[7] gives qualitative estimates of the spatial extent of turbulence and of the voltage and relaxation time for the conduction and dielectric regimes of the relaxation of turbulence after switching off the electric field.

While the theories of Carroll,[5] Ben-Abraham,[6] and Pikin[7] represent major excursions into the realm of nonlinear descriptions of nematodynamics,

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they fail to present a comprehensive theory and formalism. Our objective will be to present such a formalism as well as discussions of some cases which illustrate the richness of such a development. The results which are given here stem from comprehensive derivations by Moritz.[8]

The analysis makes use of the standard geometry,[2] where the applied electric field $\mathbf{E}_0$ is along the z-axis, the applied magnetic field $\mathbf{H}_0$ is along the x-axis, and the angle between the director $\mathbf{n}$ and the x-axis is θ.

We assume that there exists no dependence on the y coordinate and that the problem is essentially two-dimensional. If we assume that there exists some kind of spatial fluctuation in the nematic that can be described in terms of a wave vector $\mathbf{q} = (q_x, 0, q_z)$, then it is possible to state that $\theta = \theta(\mathbf{q}\cdot\mathbf{r})$. It is, therefore, also reasonable to use in the derivation the assumption that all field quantities, such as the velocity $\mathbf{v}$, and the induced electric and magnetic fields, also depend on $\mathbf{q}\cdot\mathbf{r}$. This enables us to define, in analogy with Penz and Ford,[3] a dispersion parameter $s = q_z/q_x$. The theory of Helfrich,[1] DuBois-Violette[2] and others are then the case of $s = 0$.

It can be seen immediately, with the use of either the implicit or composite function theorems, that whenever $Q = Q(\mathbf{q}\cdot\mathbf{r})$ represents a field variable such as θ or the components of $\mathbf{E}$, $\mathbf{H}$, or $\mathbf{v}$, that the following relation is true

$$\partial^n Q/\partial z^n = s^n \partial^n Q/\partial x^n \tag{1}$$

Eq. (1) allows us to reduce a two-dimensional theory to that of one dimension and a parameter. It should be pointed out that the value of s is not being fixed here, so initial alignments varying from $s = 0$ to $s = \infty$ can be treated with equal ease in our formulation.

It can be shown[8–11] that to construct a comprehensive formalism one needs to discuss only four equations, namely, the two components of the equation of motion, the torque balance equation, and the charge balance equation. The continuum model is assumed together with incompressibility and a single Frank curvature elastic constant. The usual[2–4] definitions of the anisotropic conductivity, dielectric, and stress tensors are utilized. The partial derivative with respect to x of the torque balance equation is obtained from first principles and then, utilizing Eq. (1), one can set up four coupled equations for $\dot{q}$, $\dot{\psi}$, F_3 and F_4 which are analogous to those of the linear problem.[2,4]

If we denote the curvature $\partial\theta/\partial x$ by ψ and allow q to represent the true charge density, then the charge continuity equation takes the form

$$0 = \dot{q} + \frac{q}{\tau} + \sigma_H\left[(\cos\theta + s\sin\theta)\frac{\partial}{\partial x}(\mathbf{m}\cdot\mathbf{E}) + (\mathbf{m}\cdot\mathbf{E})(s\cos\theta - \sin\theta)\Psi\right] \tag{2}$$

and the x derivative of the balance of torque equation becomes

$$0 = \dot{\psi} - \frac{K}{\gamma_1}\left\{[s^2 + (1 - s^2)\cos^2\theta]\frac{\partial^2\psi}{\partial x^2} + (1 - s^2)\cos 2\theta\psi^3\right\}$$
$$- \frac{1 + s^2}{2}\frac{\partial^2 V_z}{\partial x^2} - \frac{\gamma_2}{2\gamma_1}\left\{[(1 + 2s - s^2)(\sin 2\theta - \cos 2\theta)]\psi\frac{\partial V_z}{\partial x}\right.$$
$$\left. + [(s^2 - 1)\cos 2\theta - 2s\sin 2\theta]\frac{\partial^2 V_z}{\partial x^2}\right\} - \frac{F_2(\theta)}{\gamma_1} \tag{3}$$

where the one constant approximation $k_{11} = k_{33} = k$ for the Frank elastic coefficients and the incompressibility condition $\nabla \cdot \mathbf{v} = 0$ have been utilized. The quantity $F_2(\theta)$ is given by

$$F_2(\theta) = \cos 2\theta\frac{\partial}{\partial x}(\varepsilon_a E_x E_z + \mu_a H_x H_z) - 2\sin 2\theta(\varepsilon_a E_x E_z + \mu_a H_z H_z)\psi$$
$$- \tfrac{1}{2}\sin 2\theta\frac{\partial}{\partial x}[\varepsilon_a(E_x^2 - E_z^2) + \mu_a(H_x^2 - H_z^2)]$$
$$- \cos 2\theta[\varepsilon_a(E_x^2 - E_z^2) + \mu_a(H_x^2 - H_z^2)]\psi \tag{4}$$

and the two force components of the equation of motion are given by

$$F_3(\theta) = A_{11}(\theta)\psi\frac{\partial V_z}{\partial x} + A_{12}(\theta)\frac{\partial^2 V_z}{\partial x^2} \tag{5a}$$

$$F_4(\theta) = A_{21}(\theta)\psi\frac{\partial V_z}{\partial x} + A_{22}(\theta)\frac{\partial^2 V_z}{\partial x^2} \tag{5b}$$

where

$$A_{11}(\theta) = \alpha_1\left[\left(\cos^2\theta + \frac{s}{2}\sin 2\theta\right)\left(\frac{1 - s^2}{2}\cos 2\theta + 2s\sin\theta\right)\right.$$
$$\left. + \left(\frac{1 - s^2}{2}\sin 2\theta - s\cos 2\theta\right)(s\cos 2\theta - \sin 2\theta)\right]$$
$$+ (\alpha_5 + \alpha_6)\left(\frac{1 - s^2}{2}\cos^2 2\theta + s\sin 2\theta\right)$$
$$+ \frac{\gamma_2}{2}[2s(1 + s^2)\sin^2\theta - \cos 2\theta] \tag{6a}$$

$$A_{12}(\theta) = \alpha_1(\cos^2\theta + \tfrac{1}{2}\sin 2\theta)\left(\frac{1-s^2}{2}\sin 2\theta - s\cos 2\theta\right)$$

$$+ \frac{\gamma_2}{4}(2s^2+1)\sin 2\theta + \frac{s}{2}(1-s^2)(\alpha_5\sin^2\theta + \alpha_6\cos^2\theta)$$

$$+ (\alpha_5+\alpha_6)\left[\frac{s}{4}(1-s^2)\sin 2\theta - s\cos^2\theta\right]$$

$$- (\alpha_3\cos^2\theta - \alpha_2\sin^2\theta)s\frac{1+s^2}{2} - \frac{\alpha_4}{2}s(s+1) \tag{6b}$$

$$A_{21}(\theta) = \alpha_1\left[\left(\frac{1-s^2}{2}\cos 2\theta + 2s\sin 2\theta\right)(\tfrac{1}{2}\sin 2\theta + s\sin^2\theta)\right.$$

$$\left. + \left(\frac{1-s^2}{2}\sin 2\theta - s\cos 2\theta\right)(\cos 2\theta + s\sin 2\theta)\right]$$

$$+ \gamma_2\left(\sin 2\theta + \frac{s^3}{2}\cos 2\theta\right) + (\alpha_5+\alpha_6)s\left(s\sin 2\theta + \frac{1-s^2}{2}\cos 2\theta\right) \tag{6c}$$

$$A_{22}(\theta) = \alpha_1\left(\frac{1-s^2}{2}\sin 2\theta - s\cos 2\theta\right)(\tfrac{1}{2}\sin 2\theta + s\sin^2\theta) + \frac{\alpha_4}{2}(1+s^2)$$

$$+ \tfrac{1}{2}(1-s^2)(\alpha_5\cos^2\theta + \alpha_6\sin^2\theta) - \frac{\gamma_2}{4}s^2\sin 2\theta$$

$$- (\alpha_2\cos^2\theta - \alpha_3\sin^2\theta)(1+s^2)$$

$$+ (\alpha_5+\alpha_6)\left[s^2\sin^2\theta + \frac{s}{4}(1-s^2)\sin 2\theta\right] \tag{6d}$$

$$F_3(\theta) = -F_x(\theta) + K\left\{\left[2 + 2s^2(s+1)\sin^2\theta + s^2\frac{(s+2)}{2}\sin 2\theta\right]\psi\frac{\partial\psi}{\partial x}\right.$$

$$\left. + [\tfrac{1}{2}s^2(2s-1)\sin 2\theta + s^2(1-s)\cos 2\theta]\psi^3\right\}$$

$$+ \gamma_2(\cos 2\theta - s\sin 2\theta)\psi\dot{\theta}$$

$$+ \left[s(\alpha_3\cos^2\theta - \alpha_2\sin^2\theta) + \frac{\gamma_2}{2}\sin 2\theta\right]\dot{\psi} \tag{7a}$$

$$F_4(\theta) = -F_z(\theta) + K\left\{\left[2\cos^2\theta + s\frac{(s+2)}{2}\sin 2\theta - s\frac{(s+1)}{2}\sin^2\theta\right]\psi\frac{\partial\psi}{\partial x}\right.$$
$$\left. + [2s^3 + (s - s^3)\cos 2\theta + \tfrac{1}{2}\sin 2\theta(2 - s)]\psi^3\right\}$$
$$+ \gamma_2(\sin 2\theta + s\cos 2\theta)\psi\dot{\theta} + \left(\alpha_3\sin^2\theta - \alpha_2\cos^2\theta - \frac{\gamma_2}{2}\sin 2\theta\right)\dot{\psi} \tag{7b}$$

F_x and F_z are the x and z components of the forces due to the applied and induced electric and magnetic fields, and will be given for a specific case later.

Equations (2, 3, 5a and 5b) constitute the complete formalism and formulation of the problem of electrohydrodynamics in nematics. In order to proceed and solve the problem, it is necessary to solve for the quantities $\psi(\partial v_z/\partial x)$ and $(\partial^2 v_z/\partial x^2)$ from Eqs. (5) and substitute them in Eq. (3) to yield a set of coupled nonlinear equations for the true charge density q and the curvature ψ together with their derivatives. These equations are analogous to those obtained by DuBois-Violette, DeGennes, and Parodi[2] and Sengupta and Saupe,[4] and they reduce to their equations when one takes the limit of s and θ approaching zero.

In order to display some of the inherent richness of the nonlinear formulation, it is necessary to make some specific assumptions concerning the fields, forces, and currents. If we take the induced electric field along the x-axis to be $E_x(\mathbf{q}\cdot\mathbf{r})$, it can be shown that $E_z = E_0 + sE_x$ will satisfy $\nabla \times \mathbf{E} = 0$ and that we need to work only with H_0 since the diamagnetic polarization is weak enough to allow us to ignore the secondary fields produced by it. If we now look at the sutuation wherein space charges arranged in layers develop until the resulting fields stop the transverse current, which must surely be true for any steady state case, we obtain $J_x = 0$, which allows us to *explicitly* write down

$$E_x = -E_0/(s + r) \tag{8}$$

where

$$r = \frac{\sigma_{xx}}{\sigma_{xz}} = 2\frac{\sigma_\perp + \sigma_a\cos^2\theta}{\sigma_a\sin 2\theta} \tag{9}$$

and the electromagnetic forces can be written as

$$F_x = -\frac{qE_0}{(r+s)} + \frac{4s\varepsilon_\perp\varepsilon_\parallel E_0^2}{(r+s)^3\varepsilon_{xz}}\cdot\frac{(r\cot 2\theta - 1)}{\sin 2\theta}(r + s - 1)\psi + \frac{\mu_a}{2}H_0^2\psi\sin 2\theta$$
$$F_z = sF_x \tag{10}$$

while the derivative of the electromagnetically induced torque is given by

$$F_2(\theta) = -\mu_a H_0^2 \cos 2\theta + \frac{\varepsilon_a E_0^2 \psi}{(s+r)} \left\{ [(1-2s)r^2 - 2r^3]\cos 2\theta + \frac{2r(1-r)}{\sin 2\theta} + \sin 2\theta[2r^2 - 2(s+3)r - 3] \right\} \tag{11}$$

Note carefully that if we had chosen the case where $D_x = 0$ instead of $J_x = 0$, then $r \to r' = \varepsilon_{xx}/\varepsilon_{xz}$. Furthermore, the two cases become equivalent for very large s.

To further reduce the mathematical complexity and obtain some novel theoretical situations which may be possible to set up experimentally we can, by taking θ to be rapidly fluctuating in space about some non-small θ_0, treat the trigonometric functions of θ as being averaged (e.g., take $\sin 2\theta = m$). Then we obtain a system, utilizing Eqs. (5) and (7) in Eqs. (2) and (3), of the form

$$\dot{q} = a_{11} q + a_{12} \psi \tag{12a}$$

$$\dot{\psi} = a_{21} q + a_{22} \psi + b_1 \frac{d^2\psi}{dx^2} + b_2 \psi \frac{d\psi}{dx} + b_3 \psi^3 \tag{12b}$$

This can be done since $d\psi/dx = (\partial\psi/\partial x)[1 + s(dz/dx)]$ and we can take the characteristic dz/dx to vanish without loss of generality. The coefficients a_{ij} and b_i are obtained by comparison with the previous equations, and depend on the specific nematic material used.

We notice three specific cases of interest:

a) $\dot{q} = \dot{\psi} = 0$

b) $\dot{\psi} = 0$ but $\dot{q} \neq 0$ and

c) $\dot{q} = 0$ but $\dot{\psi} \neq 0$.

For case a) we find for the case of a complete steady state in which no quantities depend on time, and for which we may substitute for ψ in terms of q from Eq. (12a) in Eq. (12b), an equation of the form

$$f_1^2 \frac{d^2\psi}{dx^2} + f_2^2 \psi \frac{d\psi}{dx} + f_3^2 \psi^3 + f_4^2 \psi = 0 \tag{13}$$

is obtained where f_i depend on a_{ij} and b_i. For example, $f_4^2 = a_{22} - a_{21}a_{12}/a_{11}$.

Through physical considerations it can be argued that one can effectively ignore the f_2 term, in which case the solution of Eq. (13) takes the form $\psi = A sn(u, k)$ where $u = x - x_0$ and x_0 is arbitrary while k is the modulus of the elliptic integral of the first kind and is determined to be $(f_4^2/f_1^2 - 1)^{1/2}$ The amplitude A is given by $A^2 = 2k^2 f_1^2/f_3^2$.

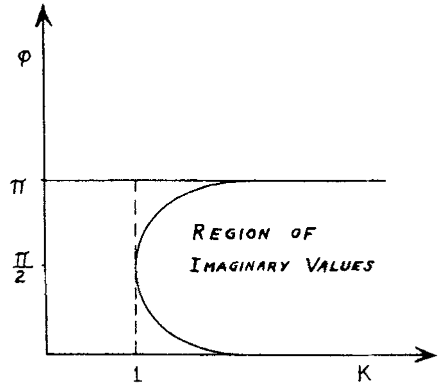

FIGURE 1 Domain of existence of solutions of Eq. (13).

The dynamics is then governed by the behaviour of the elliptic function of Jacobi $sn(u, k)$, and the region of existence of real solutions, shown in Figure 1, is then given by the condition $f_4^2/f_1^2 < 1 + \csc \phi$, where $\phi = \arcsin[sn(u, k)]$. The spatial fluctuations are seen to be doubly periodic functions which reduce to the usual trigonometric functions when k is small. To recover the full two-dimensional solution one merely needs to apply the relation (1). It should be pointed out that this analysis shows that there exists, without any a priori assumptions, domains of periodicity. But also for the same price, there are domains of instability when k takes on imaginary values. Initial calculations demonstrate that the condition on k is equivalent to threshold voltage conditions and will yield new types of instabilities. It clearly yields the usual condition for the Williams domains.

Other properties of the system for the case of externally applied forcing terms show the possibility of existence of nonlinear jump resonance phenomena for separation of domain lines as well as intensity of domain lines when the applied force is spatially periodic. This could never be obtained in a linear theory.

Case b) is a more complicated case still under investigation. The dynamics can be described by the majorante equation

$$\frac{dq}{dt} = v_1 q + (v_2 q - u_3)^{1/3} + u_4 \tag{14}$$

The phase plane diagram for this equation is given in Figure 2. The shape of the trajectory in the phase plane suggests strong correlation with the experimental curves of voltage vs. frequency.[2] We note that in the linear theories[2,4] $\theta = 0$ whereas θ is assumed constant here. We also note that our theory contains the linear terms utilized previously plus nonlinearities which modify the linear results.

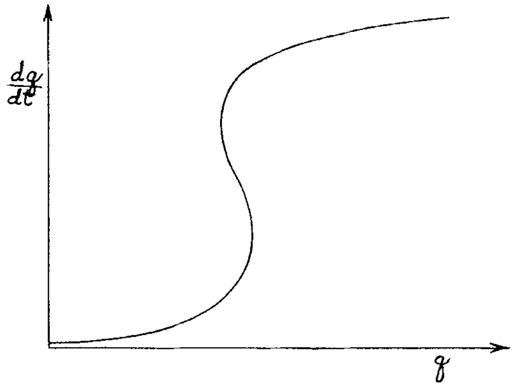

FIGURE 2 Phase plane diagram for Eq. (14).

The third and final case is the most interesting one. Eqs. (12) reduce, in this case, to $\dot{\psi} = c_1\psi + c_2\psi^3$ which is formally identical to the Landau equation[12] for the amplitudes of the velocity components of fluids near turbulence. There is strong reason to believe that the dielectric regime of dynamic scattering is described by this equation since, in rapid turbulence, the charge density does not have time to follow the fields (if they are oscillating) and in general q is averaged out to zero. This agrees with the assumptions or rather the description of this case.

In the nonlinear formulation of the electrohydrodynamic problem, nonlinearities in both the elastic and viscous portions of the nematic stress tensor might be included. The theory of quadratic nonlinearities in the viscous stress tensor has been formulated[13] and could have been included here together with nonlinearities in the elastic stress tensor. These terms are, however, neglected since they are expected to be small for velocity gradients encountered experimentally and since the coefficients have not yet been measured experimentally.

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