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Short Communication

The Maxwell stress and theoretical models of electric field effects in nematics

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Abstract. — This communication seeks to analyze the expression of the body force per unit volume arising out of the Maxwell stress caused by the application of an external electric field (E). Despite some assumptions which have been made in the derivation it appears that in addition to the term describing the effect of the electric field on the free charge density, the term corresponding to spatial variations of the electric field may also be important, especially in mathematical models attempting non-linear analyses. In cholesterics a body force caused by flexoelectricity may become important even for linear perturbations.

1. Introduction.

Owing to their anisotropic physical properties nematic and cholesteric liquid crystals exhibit a variety of interesting effects under the action of externally applied electric (E) and magnetic (H) fields. A large body of experimental observations has been explained satisfactorily on the basis of the continuum theory [1-3] for incompressible nematics by incorporating the Maxwell’s electromagnetic equations (see, for instance, [4]). One of the fascinating features of these effects is that the manner in which the external fields are imposed on the sample has an important bearing on the subsequent behaviour of the system.

When a static magnetic field H is applied normal to the uniformly aligned director, n₀, of a nematic sample a homogeneous deformation of the director field results as H is gradually raised above a well defined Fréedericksz threshold, provided that the diamagnetic susceptibility anisotropy (χₘ) is positive. Increase of H above the Fréedericksz threshold only increases the amount of homogeneous distortion in the sample; when H is very high the director field, n, in a large portion of the sample will align parallel to H.

If, on the other hand, a strong H is suddenly applied normal to n₀ the result is found to be somewhat different [5-7]. Before n assumes the equilibrium configuration determined by the
magnetic field, transient periodic dissipative structures (TPDS) are formed with wavelength of the same order as the sample thickness. The direction of periodicity of these convective structures is generally in the \((\mathbf{n}_0, \mathbf{H})\) plane. A qualitative explanation of TPDS has been given [6] on the basis of the non-stochastic continuum theory and extended by several investigators to various Fréedericksz geometries and to different varieties of nematics (see [7] and references therein). Attempts have also been made to generalize these calculations by including the effects of thermal noise [8, 9].

As the diamagnetic susceptibilities are very small, an impressed magnetic field is hardly distorted by spatial gradients of the director field. Because of this reason \(\mathbf{H}\) only contributes a torque on the director field and does not cause a significant body force affecting the law of balance of linear momentum.

The effect of \(\mathbf{E}\) is more complex. Due to the substantial dielectric constants and dielectric anisotropy \((\epsilon_A)\) of nematics, the electric field inside a nematic sample can get appreciably altered due to spatial changes in the dielectric constant arising from spatial gradients of \(\mathbf{n}\) [10]. In many situations the flexoelectric polarization [11, 10] also becomes an important factor which determines the equilibrium director configuration. In the presence of ionic impurities the electrical conductivity anisotropy \((\sigma_A)\) also enters the picture. If \(\sigma_A\) and \(\epsilon_A\) take suitable values the application of dc and ac electric fields leads to a variety of electrohydrodynamic instabilities (EHDI). The literature on EHDI is vast and the interested reader is referred to reviews [12] on the subject (see also [13] and references therein). We shall concentrate on only those studies which have a bearing on the subject of this communication.

The occurrence of a class of EHDI, known as Williams domains [1-3, 12] has been well understood on the basis of the Carr-Helfrich mechanism [14, 15]. According to this picture, free charge of opposite signs gets separated due to the electrical conductivity anisotropy under a fluctuation of the director field in the presence of \(\mathbf{E}\). The resulting destabilizing force exerted by the electric field on the free charge density \((\rho_e)\) ultimately leads to the occurrence of EHDI when the applied field (or voltage, as the case may be) exceeds a threshold; the wavevector of periodicity at the threshold can also be calculated.

The sudden application of a strong \(\mathbf{E}\) normal to \(\mathbf{n}_0\) does also lead to the observation of TPDS. In thick samples the periodic convective structures have features [16] similar to those caused by a magnetic field. In thin samples, however, there arise certain qualitative differences. It is found [17] that the direction of periodicity of TPDS is normal to the \((\mathbf{n}_0, \mathbf{E})\) plane when the nematic has high positive dielectric anisotropy. It appears [17, 18] that initially a homogeneous distortion develops in the \((\mathbf{n}_0, \mathbf{E})\) plane; subsequently this deformation suffers an instability due to perturbations whose direction of periodicity is normal to the \((\mathbf{n}_0, \mathbf{E})\) plane. The theoretical model [18, 20] is an extension of the one [6] developed to study the case of \(\mathbf{H}\).

Most of the theoretical work on the determination of the EHDI threshold has been done under the linear approximation. In this approach, the perturbations in the director, the velocity and the electric field are assumed to be small quantities of the first order; the space charge density also falls in this category. The governing equations are linearized with respect to the perturbations after relevant terms corresponding to the external torques and forces are added. The resulting equations represent an eigen value problem in which the threshold (field or voltage) is determined as the lowest eigen value associated with a certain wavevector of periodicity. In many of these efforts the main destabilizing force has been assumed to be \(\rho_e \mathbf{E}\).

Calculations have also been reported [13, 18-21] on non-linear effects above the EHDI threshold. A portion of this work [18, 20] also deals with the non-linear aspects of \(\mathbf{E}\) induced TPDS. In all these cases the body force due to the electric field appears to have been taken to be \(\rho_e \mathbf{E}\).

Magnetic and electric field effects have also been studied in the case of cholesteric (or chiral nematic) liquid crystals. The theoretical predictions made on the basis of the linear approxi-
mation [22, 23] are in good qualitative agreement with experimental results [24].

As mentioned already, the effects of external electric fields are incorporated into the continuum theory by taking into account the Maxwell's equations [4] as also the relevant boundary conditions. According to the electromagnetic theory the net force exerted on unit volume of a dielectric medium due to an applied electric field, \(f_i\), is obtained as the divergence of the relevant part of the Maxwell stress tensor \(T_{ij}\). Interestingly, expressions for the Maxwell stress have been proposed earlier in the literature [25, 26] in the case of EHD of nematics. But in these works the effects of director gradients and spatial variations of the electric field do not appear to have been taken into account.

Suppose we consider \(T_{ij}\) for an isotropic fluid dielectric and generalize it to the case of an anisotropic fluid like a nematic. It is clear that in the presence of director gradients in a nematic sample, the local permittivity can vary from point to point; in addition there can exist the effect of the flexoelectric polarization. This can be expected to cause spatial inhomogeneities in the electric field inside the fluid. If we now derive an expression for the ponderomotive force as the divergence of \(T_{ij}\), this force will have two parts. One is the familiar term \(\rho_s E\) mentioned already, this contribution exists only in the presence of free charge density. The other part is an inner product of the electric induction \(\mathbf{D}\) and the spatial gradients of \(\mathbf{E}\); this contribution can exist even in the absence of free charge and can be caused by the variations in the dielectric tensor arising from director gradients.

It is not the objective of this communication to solve specific problems related to electric field effects in nematics and cholesterics - especially the computer intensive solutions for nonlinear perturbations above threshold; these are best left to the experts who are already working in the field. The aim of the present effort is to find the external body force per unit volume exerted on a nematic or cholesteric sample by \(\mathbf{E}\) by considering linear and non-linear approximations. With this in mind a brief derivation of the body force density due to an electric field is given in section 2. It is seen that a combination of approaches given in [1] and [4] indicates the direction in which the desired result can be sought. Section 3 deals with evaluating the additional body force which might arise from inhomogeneities in the electric field. In section 4 some of the qualitative conclusions are given and the limitations of the present work are stated.

2. The equations of linear momentum balance and the body force caused by an electric field.

The electromagnetic theory can be applied to a fluid dielectric ([4] Sect. 15) to obtain an expression for the body force per unit volume, \(f^\parallel\), caused by applying an electric field produced by charged conductors maintained at constant potential,

\[
f^\parallel_k = T^\parallel_{jk,j}; \quad T^\parallel_{jk} = -P_M \delta_{kj} + (D_j E_k / 4\pi); \quad \epsilon_{jk} = \epsilon_{\delta_{jk}}; \quad \epsilon_{jk} = \epsilon \delta_{jk}; \quad (1)
\]

where \(\epsilon\) is the dielectric constant of the isotropic fluid and \(P_M\) a pressure which can be absorbed into the hydrostatic pressure, \(p\), if the fluid is incompressible; a subscripted comma denotes partial differentiation w.r.t. cartesian coordinates \(x_k\). This expression is derived by considering a small displacement \(u_i(x_k)\) of volume elements of the medium such that each volume element carries its electric potential to its new position. The net change in free energy of a given volume element caused by the displacement can be expressed as the contracted product of the stress tensor \(T_{jk}^\parallel\) and the displacement gradient \(u_{k,j}\) evaluated at that point.

The above expression cannot be conveniently used for nematics. This is because nematics have anisotropic dielectric properties with the dielectric tensor being given by

\[
\epsilon_{jk} = \epsilon_\perp \delta_{jk} + \epsilon_A n_j n_k; \quad \epsilon_A = \epsilon_\parallel - \epsilon_\perp \quad (2)
\]
where \( \epsilon_\perp, \epsilon_\parallel \) are, respectively, the dielectric constants normal to and parallel to \( \mathbf{n} \). In addition, due to spatial gradients of \( \mathbf{n} \), the flexoelectric polarization

\[
P_j = \varepsilon_1 n_j n_{k,k} + \varepsilon_3 n_k n_{j,k}
\]

(3)
can come into the picture with \( \varepsilon_1, \varepsilon_3 \) being the flexoelectric constants. Then the electric induction in a nematic becomes

\[
D_j = \varepsilon_{jk} E_k + 4\pi P_j.
\]

(4)

It is known from the continuum theory [1-3] that in the presence of director gradients the nematic experiences a body force density of curvature elastic origin,

\[
f_j^o = \sum_{k,j,k}^o; \sum_{k,j}^o = \left( \partial W / \partial n_{i,k} \right) n_{i,j};
\]

\[
W = \frac{1}{2} K_1 (\mathbf{n} \cdot \mathbf{n})^2 + \frac{1}{2} K_2 [\mathbf{n} \cdot (\nabla \times \mathbf{n})]^2 + \frac{1}{2} K_3 [(\mathbf{n} \cdot \nabla) \mathbf{n}]^2
\]

(5)

where \( K_1, K_2, K_3 \) are the curvature elastic constants and \( W \) the Oseen - Frank elastic energy density without the surface terms; for a cholesteric a term \( [k_2 \mathbf{n} \cdot (\nabla \times \mathbf{n})] \) is added to \( W \).

The expression for \( \sum_{k,j}^o \) has been derived in [1] (Sect. 3.5.2) by a process analogous to that used in [4] for deriving \( T_{jk}^f \). In the absence of an electric field, a displacement \( u_j(x_k) \) is assumed to be given to a nematic such that the director field associated with each volume element is carried to its new position. The associated change in the free energy for a given volume element \( dV \) is written as \( \delta W = \sum_{k,j}^o u_{j,k} dV \) and this immediately leads to the expression for \( \sum_{k,j}^o \) (5).

The law of momentum balance for a nematic can be written, in the most general case, as

\[
\sum_{k,j,k}^o \sum_{k,j,k}^o + f_j - \rho (d v_j / d t) = p_j
\]

(6)

where \( \sum_{k,j}^o \) is the viscous stress which depends on five viscosity coefficients, \( v_j \) the velocity and \( \rho \) the density; \( f_j \) is the external body force density caused by external fields such as electric field, gravitational field etc. As \( p \) is indeterminate for an incompressible nematic, the curl of (6) can be taken to obtain two independent equations which are supplemented by the condition of incompressibility \( (u_{k,k} = 0) \). The present task is to find an expression for \( f_j \) in (6) due to an external electric field by combining the approaches of [4] and [1].

When the electric field is caused by charged conductors which are maintained at constant potential the total free energy can be written as ([4] Sect. 13)

\[
W^T = W + W^E; \quad W^E = -(\varepsilon_{jk} E_j E_k / 8\pi) - P_j E_j;
\]

(7)
it is assumed that \( \nabla \times \mathbf{E} = 0 \). Under the action of the linear displacement \( u_j(x_k) \), a volume element at \( x_k \) goes to \( x_k' \) such that

\[
x_k' = x_k + u_k; \quad n_i(x_k') = n_i(x_k); \quad \psi_i(x_k') = \psi_i(x_k);
\]

(8)

where \( \psi \) is the electric potential and the primed quantities denote values at the new positions. As the fluid is incompressible the volume element \( dV \) does not change under the displacement. Remembering that \( \mathbf{E} = -\nabla \psi \) and following [1], the change in director gradient, \( \delta (n_{i,k}) \), and that in the electric field, \( \delta E_k \), caused by the displacement can be written as

\[
\delta (n_{i,k}) = -n_{i,j} u_{j,k}; \quad \delta E_k = -E_j u_{j,k}.
\]

(9)
It should be noted that while \( \epsilon_{jk} \) is a function of \( n_i \), \( P_j \) is dependent on \( n_p \) and \( n_{i,k} \). The change in \( P_m \) arising out of the displacement is \( \delta P_m = (\partial P_m / \partial n_{i,k}) \delta (n_{i,k}) \). Then the total change in \( W^E \) caused by (8) becomes

\[
\delta W^E = -(\epsilon_{jk} E_j \delta E_k / 4\pi) - E_m \delta P_m - P_k \delta E_k.
\]

(10)

Now \( \delta W^E \) is equated to \( T_{kj} u_{j,k} \) and using (4) an expression for the electric stress is obtained:

\[
T_{kj} = (D_k E_j / 4\pi) + E_m n_{i,j} (\partial P_m / \partial n_{i,k}).
\]

(11)

A hydrostatic pressure which arises out of the constraint of incompressibility has been left out of (11); this can be absorbed into \( p \) without loss of generality. It will be seen that if the flexoelectric polarization is neglected then (11) reduces to the relevant part of the expression given in [25, 26]. By taking the divergence of \( T_{kj} \) and utilizing the relation \( D_k,k = 4\pi \rho_e \), the external body force density can be written as

\[
f_j = f_j^{(1)} + f_j^{(2)} + f_j^{(3)}; \quad f_j^{(1)} = \rho_e E_j; \quad f_j^{(2)} = \epsilon_{km} E_m E_{j,k} / 4\pi;
\]

\[
f_j^{(3)} = P_k E_{j,k} + [E_m n_{i,j} (\partial P_m / \partial n_{i,k})]_k.
\]

(12)

By substituting for \( f_j \) into (6) and taking the curl to eliminate \( p \), the contribution due to \( f_j \) can be written as

\[
e_{p,q} f_{j,q} = e_{p,q} \rho_e, q E_j + M_p; \quad M_p = e_{p,q} [f_j^{(2)} + f_j^{(3)}]
\]

(13)

where use has been made of \( \nabla \times \mathbf{E} = 0 \). The aim of this communication is to evaluate \( \mathbf{M} \) under different approximations.

3. \( \mathbf{M} \) in first and second order approximations.

Let the unperturbed director alignment be uniformly twisted with the helical axis along \( z \) which is also the direction of \( \mathbf{E} \),

\[
\mathbf{n}_o = (C, S, 0); \quad \mathbf{E}_o = (0, 0, E_{z0}); \quad S = \sin(Qz); \quad C = \cos(Qz);
\]

(14)

\( Q \) is the wavevector of the ground state which is zero for a nematic. In the most general case, after an instability sets in we can write the director and electric fields in the form

\[
\mathbf{n} = (\cos \theta \cos \phi, \cos \theta \sin \phi, \sin \theta); \quad \mathbf{E} = (E_x, E_y, E_{z0} + E_z)
\]

(15)

where \( \theta, \phi, E_x, E_y, E_z \) are functions of \( x, y, z \) and (time) \( t \) [27]. In the zeroth order approximation (14) where there are no perturbations \( M_p = 0 \). Nematics are studied first.

In a first order approximation we assume that \( \theta, \phi, E_x, E_y, E_z \) and their spatial gradients are small. Then we ignore products of these quantities and also products of their gradients. It is then found that \( f_j^{(2)} \approx \epsilon_{1z} E_{z0} E_{j,z} / 4\pi \) and its curl vanishes as the electric field is derivable from a potential. Similarly the term \( P_k E_{j,k} \) in \( f_j^{(3)} \) is neglected to first order as both factors are first order quantities. The remaining term yields \( f_j^{(3)} \approx \epsilon_{3z} E_{z0} \theta_{j,z} \); again its curl vanishes. Hence when the perturbations are of first order, \( \mathbf{M} \) can be neglected.
Clearly, $f_j$ depends on second order derivatives of the director orientation with flexoelectric coefficients. For nonlinear perturbations these terms will naturally modify similar terms arising out of $f_j^2$ (5). This is analogous to the modification of elastic torques by the presence of flexoelectricity [10, 28] and becomes especially clear if the complete form for $f_j$ is worked out in tensor notation. Unfortunately the expressions are extremely complicated and it does not seem to be worth including them in this communication which does not set out to solve problems. In order to get an idea of the nonlinear form of M, $f_j$ is worked out below for second order perturbations including only the dielectric terms.

When the perturbations are assumed to be of second order in magnitude we retain all second order products of the perturbations and their derivatives. Then we find that

$$M_x = \varepsilon_A E_{10} (\theta_{zz} E_{yz,x} - \theta_{yy} E_{xz,x}) / 4\pi + \ldots$$
$$M_y = \varepsilon_A E_{20} (\theta_{zx} E_{zx,y} - \theta_{zz} E_{zx,x}) / 4\pi + \ldots$$
$$M_z = \varepsilon_A E_{20} (\theta_{xy} E_{yy,x} - \theta_{yx} E_{xy,x}) / 4\pi + \ldots$$

(16)

Thus in a second order approximation, M (13) does not vanish identically. In the general case where the direction of periodicity of rolls lies in the $xy$ plane [13, 19] all three components of M (13) will remain. Even in the particular case of periodicity along $x$, $M_y$ will exist; this is obtained by eliminating the pressure from the $x$ and $z$ components of (6). The dielectric contribution can exist even in a situation where the effect of flexoelectricity is not important. For example in a nonlinear analysis of electric field induced TPDS in materials like 5CB [17, 18, 20] which have high dielectric anisotropy one can expect significant contributions from the dielectric part of M.

Some of the above conclusions are valid for a cholesteric ($Q \neq 0$). To first order, $e_{pji} f_j^{(2)} \approx 0$; $P_k E_{j,k} \approx 0$. But the remaining term in $f_j^{(3)}$ yields a nonvanishing contribution even to first order,

$$f_x^{(3)} = R_x; \quad f_y^{(3)} = R_y; \quad R_k = e_3 E_{20} (C\theta_{zx} + S\theta_{zy})_k;$$
$$f_z^{(3)} = R_z + Q (e_1 + e_3) [E_{x,y} (C^2 - S^2) + SC (E_{y,y} - E_{x,x})] + (e_1 - e_3) Q E_{20} (C\theta_{zy} - S\theta_{zx});$$

(17)

As $\nabla \times R = 0$, $M_z = 0$, but $M_x$, $M_y$ will survive; these will vanish identically if $Q = 0$, as they should for a nematic.

It must be remembered, however, that the analysis in [23] will remain unaffected as flexoelectricity has not been included there. The expressions for $M_k$ can be written down for nonlinear perturbations but these are far more complicated than those given in (16).


The present work seeks to analyze the body force density due to external electric and magnetic fields in nematics and cholesterics. Due to its negligible interaction with materials a magnetic field does not give rise to a significant body force. On the other hand an electric field does give rise to a body force density $f_i$ which is the divergence of the relevant part of the Maxwell stress; in this work, $f_i$ is derived by using the approaches adopted in [4] and [1]. $f_i$ has three parts. The first part arises due to the effect of the electric field on the space charge density. The second part owes its origin to spatial gradients in E and can exist even in the absence of the flexoelectric polarization. The third part originates from director gradients and
their interaction with the electric field via the flexoelectric polarization; this contribution may significantly modify the curvature elastic body force $f_{ij}^e$ just as, in the torque equations, the presence of flexoelectricity can lead to a redefinition of elastic torques \cite{10,28}. The first part of $f_{ij}$ has been taken into account in linear and nonlinear analyses of EHDI and also in the linear analysis of electric field induced TPDS; in nematics the remaining parts of $f_{ij}$ get eliminated with $p$. But in cholesterics the flexoelectric part of $f_{ij}$ makes a nonvanishing contribution even for linear perturbations. In nonlinear analyses of EHDI and electric field induced TPDS both parts of $f_{ij}$ have to be taken into account for a full theoretical description of the phenomenon. It appears that this has not so far been done in nonlinear calculations on EHDI in nematics \cite{21,13,19}.

The question does arise as to how important the additional terms really are. Before attempting an answer, it must be emphasized that a mere derivation of the expression of $f_{ij}$ is not sufficient. As the dynamics of nematics and cholesterics is characterized by a coupling between the director and velocity fields, a complete understanding of the effects of the additional terms will be known only by solving all the governing equations in a given situation. This reduces to a numerical solution of several coupled nonlinear partial differential equations and this falls outside the scope of the present work.

No claim is made that the expression for $T_{kj}$ \cite{11} is the final one. It must be remembered that there are some assumptions involved in the derivation. For instance, the expression for the free energy density \cite{7} is derived under the assumption that there are no free charges in the sample \cite{4} Sect. 10); in the presence of free charges the effects of electrical conductivity may become important. Then again, only the bulk free energy density has been used. It is known \cite{1-3} that many interesting effects occur due to weak anchoring of the director field at the sample boundaries; this is taken into account by adding the surface free energy to the volume contribution. It must also be remarked that in the present work the exact shape of the sample has not been taken into account. When the sample assumes a specific shape and the potential difference applied to a nematic is property calculated \cite{10,28} it is found that the flexoelectric polarization can affect both the bulk torque and the surface torque on the director field; this can become important when the anchoring is weak. For a more rigorous description the full expression of the free energy may have to be taken. Another aspect of flexoelectricity is that in many static situations involving weak anchoring it may lead to polar electric effects \cite{29} and Refs. therein); this means that when the sign of the applied (dc) electric field is reversed the net electric torque can change even in magnitude. One can see from \cite{11} that in the presence of flexoelectricity a similar effect may also be expected in the ponderomotive force.

Lastly, the present work has considered only incompressible nematics and cholesterics. In the more general case of a compressible fluid, it will be necessary to include the remaining terms which give, for instance, variation of dielectric constants with density. Such a general formulation may be useful for studying electric field effects on propagation of compressional waves in the medium.

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References

[27] As the curl of the electric field is assumed to vanish we can get an equivalent description [13] by assuming that $E_0 = \psi_0 z$ and $(E_x, E_y, E_z) = (\psi''_x, \psi''_y, \psi''_z)$ where $\psi''$ is the perturbation imposed on the equilibrium electric potential $\psi_0$.