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On the Ability of Homogeneously Aligned Nematic Mesophases with a Positive Dielectric Anisotropy to Exhibit Williams Domains as a Threshold Effect

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The ability of homogeneously aligned nematics with a positive dielectric anisotropy to exhibit Williams Domain as a threshold effect is numerically investigated. A simplified two-dimensional model which has, already, given satisfactory results in the negative dielectric anisotropy case is used. The frequency dependence of the threshold fields obtained for materials with small dielectric anisotropies is in satisfactory qualitative agreement with the known experiments. The frequency dependence of the threshold spatial wavenumbers is calculated. In accordance with earlier work, a positive dielectric anisotropy nematic which normally does not exhibit Williams Domains, does so under the action of a stabilizing magnetic field. This effect is confirmed and the frequency dependence of the threshold fields as well as the associated threshold spatial wavenumbers, for various values of the stabilizing field, are calculated. In addition, the model predicts the existence of a class of positive dielectric anisotropy nematics which do not form Williams Domains whatever the applied magnetic field is. This suggests a distinction between three classes of positive dielectric anisotropy nematics.

I INTRODUCTION

The problem of the electrohydrodynamic (EHD) excitation of nematic mesophases (NM) with a positive dielectric anisotropy (PDA) has been a subject of a lesser interest than that of the negative dielectric anisotropy (DNA) ones. The preceding work[1–11] suggests that when a homogeneously aligned nematic layer with a PDA is excited with an audio frequency electric field, normal to the layer, at sufficiently high frequencies, and above a certain threshold voltage, independent of the frequency, an one-dimensional re-orientation of the director field (Freedericsz Deformation (FD)) is observed.[1–5]

On the other hand, the possibility exists for some PDA nematics to exhibit Williams Domains (WD) at lower frequencies and with a lower threshold voltage,[2,3,6–8] which, however, is an increasing function of the frequency becoming, at a certain value of the frequency, equal to the FD threshold.[2,3] In the rest of the frequency range reorientation, as a threshold effect, is observed.

It appears that the main question to be answered concerns the ability of a PDA nematic to form WD, and if so, to predict quantitatively the frequency dependence of the threshold voltage and the associated spatial wavenumber. The question has been discussed rather qualitatively[3,10,11] in terms of the one-dimensional model of the EHD excitation.[12–14] It is already realized, however, that the answer should be given in terms of a two-dimensional model.[3] The only quantitative results are those of Penz,[15] who worked out a rigorous two-dimensional model in the case of d.c. excitation. His results describe the influence of the dielectric anisotropy to the ability of a PDA nematic to form WD.

This article presents the results of a recently reported method[16] to predict EHD threshold fields and the associated spatial wavenumbers, when it is applied to the case of PDA nematics. The method is based on a simplified two-dimensional model and so is not rigorous in the sense of Penz's model.[17] Nevertheless, the degree of agreement of the results of the method[16,18] with the already known experiments,[19,20,24] in the case of NDA, justifies the present application, as far as the qualitative features of the effects involved are under study.

II THE METHOD

The EHD excitation of a homogeneously aligned nematic layer by an electric field normal to it, is described by the coupled system of differential equations[13,14,16]

$$\dot{q} + a_1 q + b_1 E \psi = 0 \tag{II.1a}$$

$$\dot{\psi} + a_2 \psi + b_2 E q = 0 \tag{II.1b}$$

where q and ψ are, respectively, the excess charge density and the director curvature along the initial orientation direction, E is the instantaneous value of the applied electric field, a_1 and a_2 are, respectively, the inverses of the charge and director relaxation time, and b_1 and b_2 are the coupling coefficients. Analytical expressions for the parameters of (II.1) are given in Appendix A. In what follows the results of the application of a square-pulsed electric field will be discussed. This saves computer time and the results obtained are qualitatively true for the sinusoidal excitation case, as well.

When a square-pulsed electric field of frequency f and amplitude E is applied to a NM layer, in order that (II.1) has a non-zero steady-state solution the following condition must hold[16]

$$(a_1 - a_2)\sinh\left(\frac{D}{4f}\right) = D\sinh\left(\frac{a_1 + a_2}{4f}\right) \tag{II.2}$$

where

$$D \equiv +\{(a_1 - a_2)^2 + 4b_1b_2E_2\}^{1/2}$$

is restricted to real values. Condition (II.2) refers to the conduction mode of EHD excitation.

Equation (II.2) defines the amplitude E of the applied electric field as an implicit function of the frequency of the field and the value k of the spatial wavenumber. Thus, for a given f, the minimum of E is the threshold field at this f whereas the corresponding k is the value of the spatial wavenumber associated with this threshold.

III THE INFLUENCE OF THE DIELECTRIC ANISOTROPY

The outlined procedure is applied, with the aid of a digital computer, to a hypothetical NM which possesses all the physical parameters of the room temperature nematic MBBA,[21–24] except of the dielectric constant ε_p, parallel to the director, which is varied as a free parameter. This is done because of the lack of any complete list of PDA nematic parameters, and this is the main reason why our results are of qualitiative value only. (For the physical parameters used see Appendix B).

Figure 1 shows a family of curves for the frequency dependence of the threshold field, where the dielectric anisotropy takes the values $\varepsilon_\alpha = 0$, 0.02, 0.05, 0.2 and 0.5. The left (curved) part of each curve corresponds to the formation of WD, and the right (horizontal) part to the FD threshold. It is evident from the figure that the WD part of the curves is relatively insensitive to the variations of ε_α, and only the frequency limit of this part at which the WD and FD thresholds become equal depends strongly on ε_α. As explained in Appendix A the plots are given in terms of the quantities E_r and f_r defined there. For the conductivity and the stabilizing magnetic field term, respectively, the values

$$\sigma_r = 250 \text{ cgs}, \ Q = 0$$

are used.

The FD threshold is determined by the condition

$$a_2 = 0 \tag{III.1}$$

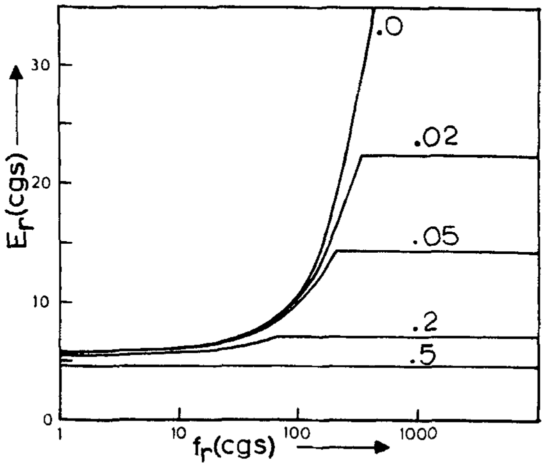

FIGURE 1 Frequency dependence of the threshold field computed for $\varepsilon_\alpha = 0$, 0.02, 0.05, 0.2, 0.5. The horizontal part of each curve corresponds to reorientation thresholds and the curved part to WD thresholds. The plots are obtained using $\sigma_r = 250$ cgs and $Q = 0$.

This follows directly from (II.1) by setting $q = 0$ and demanding for the director field to have a non-zero deformation. Relation (III.1) gives, after some algebra, and using the definitions of Appendix A, the well known formula for the FD threshold voltage[3]

$$V_{FD} = 2\pi(\pi K_{11} + \chi_a H^2 L^2)^{1/2}/\varepsilon_\alpha^{1/2}$$

Figure 2 gives the respective family of curves for the threshold spatial wave numbers expressed in units of m. The case $\varepsilon_\alpha = 0.5$ is trivial giving $k = 0$

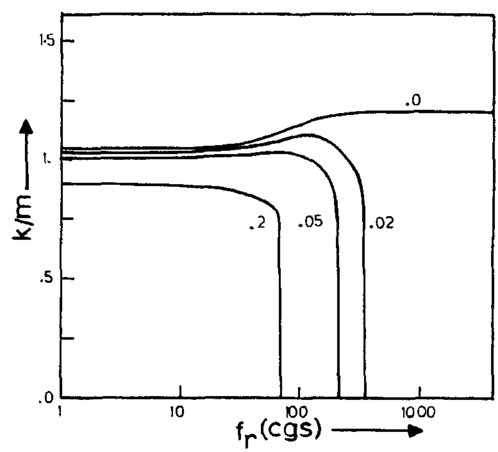

FIGURE 2 Frequency dependence of the threshold DSW associated with the curves of Figure 1. The plot of the $\varepsilon_\alpha = 0.5$ case coincides with the frequency axis.

for all frequencies. All the other curves, except of that for which $\varepsilon_\alpha = 0.5$, exhibit an abrupt fall of the wavenumber near the frequency at which WD and FD thresholds become equal. Depending on the value of the dielectric anisotropy, the curves may or may not exhibit a slight maximum.

The curves of Figure 1 are in qualitative agreement with the experimental ones of Refs. 2 and 3. No experimental work exists, however, corresponding to the results of Figure 2.

IV THE INFLUENCE OF THE MAGNETIC FIELD

As reported and discussed in Ref. 11, the application of a stabilizing magnetic field (magnetic field parallel to the initial orientation direction) may cause a PDA nematic that normally does not form WD, to do so when the magnetic field reaches a certain value, which depends on the excitation conditions. The effect is explained, within the framework of the Carr–Helfrich model, in terms of the competition of the various torques inside the NM.

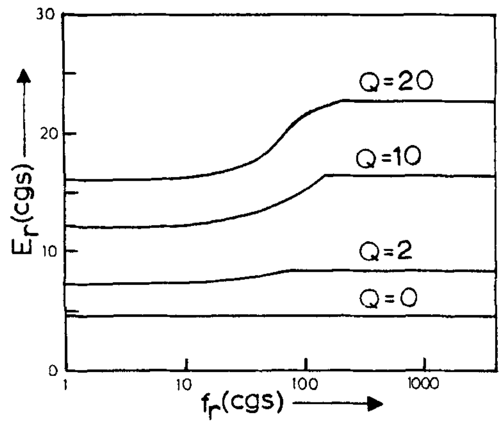

FIGURE 3 Frequency dependence of the threshold field computed for $\varepsilon_\alpha = 0.5$ and for $Q = 0, 2, 10, 20$. (In the cgs system of units used here Q is a dimensionless quantity). The plot corresponds to a moderately positive nematic (see text). Again $\sigma_r = 250$ cgs.

Our calculations confirm the effect. A family of $E_r - f_r$ curves is plotted in Figure 3, using $\varepsilon_\alpha = 0.5$ and $\sigma_r = 250$ cgs, and letting the stabilizing magnetic field term taking the values $Q = 0, 2, 10$ and 20. As is evident from the figure the difference between WD and FD thresholds at some fixed frequency, as well as, the frequency range at which WD are observed, increase monotonically with Q. The corresponding family of wavenumber curves is plotted in Figure 4. A general increase of the threshold spatial

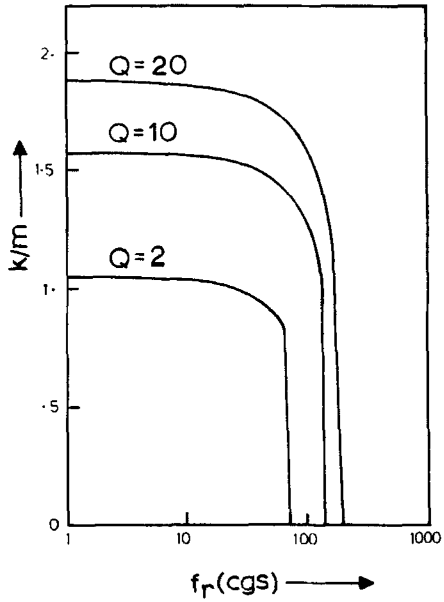

FIGURE 4 Frequency dependence of the threshold DSW associated with the curves of Figure 3. The plot of the $Q = 0$ case coincides with the frequency axis.

wavenumbers with Q is apparent. Again, the case $Q = 0$ is trivial giving $k = 0$ in the entire frequency range.

V MAGNETIC FIELD VERSUS DIELECTRIC ANISOTROPY

In terms of the simplified two-dimensional model used here, a criterion, concerning the ability of a NM to form WD under giving stabilization conditions, may be found.

When the frequency of the applied field is small enough, the time derivatives in (II.1) may be ignored, so that the (d.c.) instability condition becomes

$$a_1 a_2 = b_1 b_2 E^2$$

which, using (A.5), becomes

$$E_r^2 = \frac{p_0}{b_1 b_1 / a_1 + p_1} \tag{V.1}$$

where p_0, p_1, a_1, b_1 and b_2 are used as they are defined in (A.11), (A.7), (A.9), (A.10) and (A.8), respectively. The threshold field is determined by the relation

$$E_{r,\mathrm{th}}^2 = \min\left\{\frac{p_0}{b_1 b_2/a_1 + p_1}\right\} \tag{V.2}$$

Now, in order that the NM *does not* form WD as a threshold effect the following condition must hold for all $S \neq 0$.

$$\frac{p_0}{(b_1 b_2/a_1) + p_1} \geq \left(\frac{p_0}{(b_1 b_2/a_1) + p_1}\right)_{S=0} = \frac{4\pi(Q + K_{11}/K_{33})}{\varepsilon_\alpha} \tag{V.3}$$

The last equality follows directly from the definitions of Appendix A. The expression in the right hand side of the inequality in (V.3) is, of course, the FD threshold field. Using, now, (A.9), (A.10), (A.11), (A.7) and (A.8), and ignoring the diffusion current term of the parameter a_1 because for a wave-numbers involved its effect is negligibly small, (V.3) gives

$$\min\left(\frac{\sigma_\alpha}{\sigma_p} + \frac{S + \sigma_n/\sigma_p}{Q + K_{11}/K_{33}} - \frac{\varepsilon^p}{\varepsilon_\alpha}\left(\frac{\varepsilon_n}{\varepsilon_p} - \frac{\sigma_n}{\sigma_p}\right)\frac{\xi\xi_\alpha}{\xi_n}\right) \geq 0 \tag{V.4}$$

where

$$\sigma_\alpha \equiv \sigma_p - \sigma_n$$
$$\xi \equiv S + 1 \tag{V.5}$$
$$\varepsilon_\alpha > 0$$

When the relation (V.3) or its equivalent (V.4) is not satisfied, the NM will exhibit WD from d.c. (i.e. very low frequency) up to some frequency limit. The relation (V.4), with the aid of a digital computer, gives the curve of Figure 5 where the relation between the dielectric anisotropy and the stabilization term Q needed for WD to be formed, is plotted. The region above the curve corresponds to WD conditions (provided the frequency is small enough), whereas the region below the curve corresponds to reorientation thresholds in the entire frequency range. As Figure 5 suggests, there exists a value $\varepsilon_{\alpha 2}$ for the dielectric anisotropy such that, for all ε_α greater than this value, there is no possibility to obtain WD no matter how large the applied magnetic field is. This fact suggests the distinction between three classes of PDA nematics. The weakly positive nematics which give WD that may be quenched with the application of a destabilizing magnetic field (magnetic field perpendicular to the nematic layer). They correspond to that part of the ε_α axis for which the curve of Figure 5 lies below the $Q = 0$ line. The moderately positive nematics which, normally, do not exhibit WD, but do so under the action of a stabilizing magnetic field. And the strongly positive nematics which do not form WD whatever the applied magnetic field is.

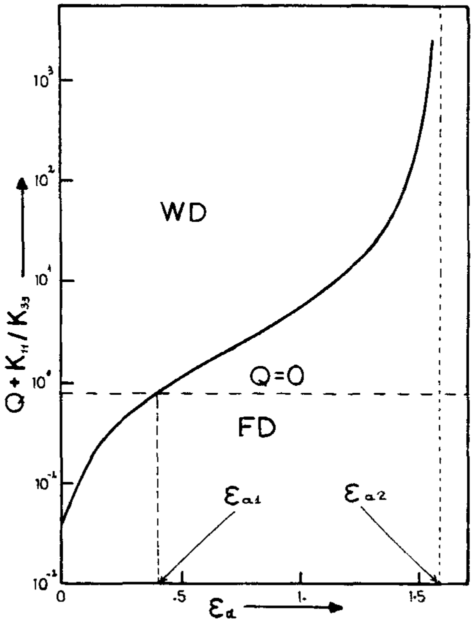

FIGURE 5 Dielectric anisotropy dependence of the minimum stabilization term Q needed for the system to form WD. The section of ε_α axis for which the curve lies below the $Q = 0$ line, corresponds to weakly positive NM. The section between $\varepsilon_{\alpha 1}$ and $\varepsilon_{\alpha 2}$ corresponds to moderately positive nematics, whereas the remaining (infinite) part to strongly positive NM.

VI HOW TO INFER STRONG POSITIVITY

Using condition (V.4), a strong positivity criterion may be found as follows:

The expression $\xi\xi_\alpha/\xi_\eta$ in (V.4), using (A.3), (A.4) and (V.5), is easily shown that increases monotonically with S, that is

$$\frac{\partial}{\partial S}\left(\frac{\xi\xi_\alpha}{\xi_\eta}\right) > 0 \tag{VI.1}$$

provided that the Leslie's viscosity coefficients[25] satisfy the following conditions

$$\alpha_2 < 0, |\alpha_3| \ll |\alpha_2| \tag{VI.1a}$$

which is the case for most nematics. Now, if we define the expression inside the brackets in (V.4) as $f(S)$, the following conditions must, simultaneously, hold, for a point on the curve of Figure 5

$$f(S) = 0, \frac{\partial f(S)}{\partial S} = 0$$

The second of them gives

$$\frac{\sigma_n/\sigma_p}{Q + K_{11}/K_{33}} = \frac{\varepsilon_p}{\varepsilon_\alpha}\left(\frac{\varepsilon_n}{\varepsilon_p} - \frac{\sigma_n}{\sigma_p}\right)\frac{\partial}{\partial S}\left(\frac{\xi\zeta_\alpha}{\xi_\eta}\right)$$

and, because of (VI.1) one must have

$$\frac{\varepsilon_n}{\varepsilon_p} - \frac{\sigma_n}{\sigma_p} > 0 \tag{VI.3}$$

It is not difficult to see that as $Q \to \infty$, the solution of (VI.2), considered as an equation in S, tends to infinity with the rate of $Q^{1/2}$ and, thus, the expression

$$\frac{S + \sigma_n/\sigma_p}{Q + K_{11}/K_{33}}$$

tends to zero as Q goes to infinity. Therefore, for $Q \to \infty$, the condition (V.4), with the help (VI.1) and (VI.3) gives

$$\frac{\sigma_\alpha}{\sigma_p} - \frac{\varepsilon_p}{\varepsilon_\alpha}\left(\frac{\varepsilon_n}{\varepsilon_p} - \frac{\sigma_n}{\sigma_p}\right)\max\left(\frac{\xi\zeta_\alpha}{\xi_\eta}\right) \geq 0 \tag{VI.4}$$

and, finally, using (A.3), (A.4), (V.5) and (VI.1) one finds

$$\frac{\varepsilon_\alpha}{\varepsilon_n} \geq \left(\frac{\sigma_n}{\sigma_\alpha} - \frac{\eta_1}{\alpha_2}\right)^{-1}$$

The strong positivity condition (VI.4), and also the condition (VI.3), may not of course be valid when restrictions (VI.1a) are not fulfilled. The condition (V.4), however, is free of any such assumption and depends only on the positivity of the dielectric anisotropy.

For the nematic parameters used, the criterion (VI.4) gives

$$\frac{\varepsilon_\alpha}{\varepsilon_n} \geq 0.30$$

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Appendix A

Let L be the thickness of the nematic layer and k the spatial wavenumber. We define the deformation ratio as

$$S \equiv \frac{k^2}{m^2}$$

where

$$m \equiv \frac{\pi}{L}$$

The parameters a_1 and b_1 are given by the relations

$$a_1 \equiv \frac{4\pi(\sigma_p S + \sigma_n)}{\varepsilon_p S + \varepsilon_n} + D_p k^2 + D_n m^2 \tag{A.1}$$

$$b_1 \equiv (\sigma_p \varepsilon_n - \varepsilon_p \sigma_n) \frac{S + 1}{\varepsilon_p S + \varepsilon_n} \tag{A.2}$$

where ε_p and ε_n are, respectively, the dielectric constant along and normal to the director, σ_p and σ_n the respective conductivities and D_p and D_n the respective mean diffusion coefficients.

Now, let $\alpha_1, \ldots, \alpha_6$ be the Leslie's viscosity coefficients[25] for an incompressible NM, for which the following relation exists[26]

$$\alpha_6 - \alpha_5 = \alpha_3 + \alpha_2$$

We define[16]

$$\eta_1 \equiv \frac{\alpha_4 + \alpha_5 - \alpha_2}{2}$$

$$\eta_2 \equiv \alpha_1 + \alpha_3 + \alpha_4 + \alpha_5$$

$$\gamma_1 \equiv \alpha_3 - \alpha_2$$

$$\gamma_2 \equiv \alpha_3 + \alpha_2$$

and also

$$\xi_\alpha \equiv \alpha_3 - \alpha_2 S \tag{A.3}$$

$$\xi_\eta \equiv \eta_1 S^2 + \eta_2 S + \eta_1 + \gamma_2$$

$$\eta \equiv \gamma_1 - \frac{\xi_\alpha^2}{\xi_\eta} \tag{A.4}$$

The parameter a_2 is defined as follows

$$a_2 \equiv p_0 - p_1 E^2 \tag{A.5}$$

where

$$p_0 \equiv \frac{K_{33}k^2 + K_{11}m^2 \pm \chi_\alpha H^2}{\eta} \tag{A.6}$$

and

$$p_1 \equiv \frac{\varepsilon_\alpha \varepsilon_n(S+1)}{4\pi(\varepsilon_p S + \varepsilon_n)\eta}, \qquad \varepsilon_\alpha \equiv \varepsilon_p - \varepsilon_n \tag{A.7}$$

where K_{11} and K_{33} are, respectively, the splay and bend curvature elastic constants of the NM, H is the applied magnetic field and χ_α is the anisotropy of the magnetic susceptibility of the NM. The sign of the magnetic field term is positive when the field is parallel to the initial orientation direction (stabilizing field), and negative when the field is perpendicular to the nematic layer (destabilizing field).

Finally, b_2 is given by

$$b_2 \equiv S\,\frac{\xi_\alpha/\xi_\eta - \varepsilon_\alpha/(\varepsilon_p S + \varepsilon_n)}{\eta} \tag{A.8}$$

Now, because of the form of the equation (II.2) the following parameters, in place of the preceding ones, may be used

$$f \to f_r \equiv \frac{f}{K_{33}m^2}$$

$$E \to E_r \equiv \frac{E}{(K_{33}m^2)^{1/2}}$$

$$H \to Q \equiv \frac{\pm\chi_\alpha H^2}{K_{33}m^2}$$

$$\sigma_p \to \sigma_r \equiv \frac{\sigma_p}{K_{33}m^2}$$

$$D_p \to D_r \equiv \frac{D_p}{K_{33}}$$

and the parameters a_1, b_1 and p_0 are redefined as follows

$$a_1 \equiv \frac{4\pi\sigma_r(S + \sigma_n/\sigma_p)}{(\varepsilon_p S + \varepsilon_n) + D_r(S + D_n/D_p)} \tag{A.9}$$

$$b_1 \equiv \frac{\sigma_r(\varepsilon_n - \varepsilon_p\sigma_n/\sigma_p)(S + 1)}{\varepsilon_p S + \varepsilon_n} \tag{A.10}$$

$$p_0 \equiv \frac{S + K_{11}/K_{33} + Q}{\eta} \tag{A.11}$$

In this way the explicit dependence of the results on L and K_{33} is supressed.

Appendix B

The physical parameters used for the calculations are as follows Viscosity coefficients:[21]

$$\alpha_1 = 0.07P, \alpha_2 = -0.78P, \alpha_3 = -0.01P, \alpha_4 = 0.83P, \alpha_5 = 0.46P.$$

Curvature elastic constants:[22]

$$K_{33} = 0.07 \times 10^{-6} \text{ dyn}, \frac{K_{11}}{K_{33}} = 0.8$$

Electrical properties:[23]

$$\varepsilon_n = 5.25, \frac{\sigma_p}{\sigma_n} = 1.5.$$

Diffusion constants:[24]

$$D_p = 0.5 \times 10^{-7} \text{ cm}^2/\text{sec}, D_n = 0.33 \times 10^{-7} \text{ cm}^2/\text{sec (estimated)}$$

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