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INTERPRETATION OF THE INSTABILITIES UNDER AC ELECTRIC FIELDS IN THICK SAMPLES OF S_c^* LIQUID CRYSTALS

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We explain by a simplified theory the existence of the undulation instability found by us in thick samples of planar oriented S_c^* liquid crystals. We derive the order of magnitude of the threshold field and explain the minimum in the frequency dependence of the unwinding critical field E_c as well.

INTRODUCTION

A number of instabilities induced by AC electric fields were found in various liquid crystal phases. In *nematic phases* at a certain threshold voltage V_c thin (in the order of sample thickness) cylindrical vortices (so called Williams domains) perturbing the initial orientation of the director form by the mechanism explained (based on the idea of Carr[1]) by Helfrich.[2] Similar instabilities were found in *cholesteric* phases[3,4,5] and explained first by Hurault.[6]

There are electrohydrodynamic instabilities in smectic phases as well and were observed[7,8,9] in *nonchiral* S_c materials, however so far the theoretical explanation is missing. In *chiral* S_c^* materials the existence of electric field induced smectic layer distortions was predicted and calculated theoretically for homeotropic cells already in 1979 by Noel A. Clark.[10]

As far as we know this prediction was verified only by us in planar S_c^* samples recently. We observed that in thick samples (the sample thickness is larger than the pitch) at electric fields larger than a threshold, E_{th}, but less than the unwinding critical field E_c, an undulation instability of the originally straight parallel stripes occurs.[11] At fields just above the threshold ($E \gtrsim E_{th}$) the distortion of the stripes was found to be sinusoidal and had a zig-zag shape at higher fields.

Furthermore, a minimum was observed in the frequency dependence of the unwinding critical field E_c. The minimum in the frequency response coincides with the characteristic frequency f_{RC} ($f_{RC} = \sigma/(2\pi\varepsilon)$, σ and ε are the average conductivity and dielectric constants respectively) of the substance. In the vicinity of this frequency the unwinding is caused by a flow suggesting that the underlying mechanism is linked to the conductivity.

The aim of this paper is to explain the existence of the measured undulation instability and the anomaly in the $E_c(f)$ function in the frame of the same theory.

I. MODEL

The background structure i.e. the configuration just below the undulation threshold is shown in Figure 1. On the basis of the model of Glogarova *et al.*[12] and our measurement on switching times of FK4[13] we assume that below the electric field $E_{th} \sim 10^5$ V/m the film consists of a sequence of parallel stripes of domains separated by dechiralization lines (instead of a uniform helical structure). In the domains the spontaneous polarization P_0 is alternatingly parallel and antiparallel to the electric field at all the frequencies. (see Figure 1) (FK4 is a room temperature binary mixture[14] made by two compounds of 4-(2′-methylbutiloxy)-phenil-4-alkiloxy-benzoate homologues (MBOPEnOBA) containing 60% by weight of the compound with $n = 8$ and 40% by weight of $n = 12$).

Due to the anisotropy of the electric conductivity this periodical structure in the z direction (z is parallel to the smectic layer normal n) yields a spatially periodic electric field component in this direction inside the sample. As a result of this component of the field, E_z, it will be energetically favourable if the polarization has a component parallel to z. For this reason a perturbation of the azimuth angle φ, and the displacement of the layers u occur. The boundary conditions for u and φ at the boundaries of the smectic monodomains are: $u = 0$ and $\varphi = 0$. On the basis of the former considerations we will examine the following perturbations:

$$\delta u(y, z) = u_0 \cdot \cos qy \cdot \cos(\pi z/d) \tag{1}$$

$$\delta \varphi(y, z) = \varphi_0 \cdot \sin qy \cdot \cos(\pi z/d) \tag{2}$$

here q is the wave number of the sinusoidal deformation, and d is the dimension of the smectic monodomains. (As mentioned in Reference 11, in the samples there were regions with diameters of about 200–300 μm which were perfectly aligned with their smectic layers in one direction, but the difference between the directions in the neighbouring regions did not exceed 5°).

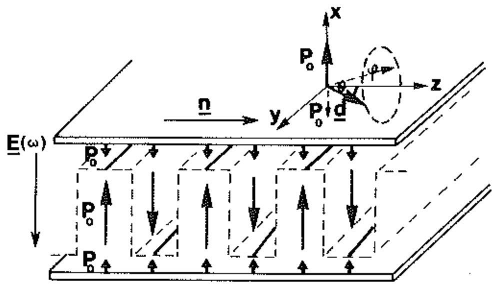

FIGURE 1 Model of the structure of thick planar S_c^* sample just below the electric field $E_{th} \sim 10^5$ V/m. The cell consists of a sequence of parallel stripes of domains (dashed line). In the domains the spontaneous polarization $\mathbf{P}_0$ is alternatingly parallel and antiparallel to the electric field $E(\omega)$ at all the frequencies. Satisfying the boundary condition, near to the bounding plates straight parallel stripes of so called dechiralization lines[12] exist in y direction (these indicated by continuous lines). The director d makes an angle θ with the smectic layer normal n. If the azimouth angle $\varphi = 0$ $\mathbf{P}_0$ points up (indicated by continuous arrow), and if $\varphi = \pi$ $\mathbf{P}_0$ points down (indicated by dashed arrow).

II. THEORY

(A) Assumptions

For simplicity we use the following approximations:

1. $E_x = E$ is constant in the sample (As it can be seen in Figure 1 near to the walls, the director must also depend on x to satisfy the correct boundary conditions. However in our approximate treatment this is not taken into account.)

2. As S_c^* is a locally biaxial system the dielectric tensor ε has three different eigenvalues ε_1, ε_2 and ε_3 with the main axes: firstly the direction of spontaneous polarization ($\mathbf{n} \times \mathbf{c}$); secondly the direction perpendicular both to the spontaneous polarization and the director ($\mathbf{d} \times (\mathbf{n} \times \mathbf{c})$); thirdly the director $\mathbf{d}$. Here $\mathbf{n}$ and $\mathbf{c}$ denote the smectic layer normal and the projection of the director on the smectic layers respectively.

The director $\mathbf{d}$ (which can be distinct from the director as defined from theory of elasticity)[18] makes an angle θ_0 with the smectic layer normal.

3. The electric conductivity tensor $\boldsymbol{\sigma}$ has three different eigenvalues (σ_1, σ_2, σ_3) as well. For symmetry reasons the first principal axis should be the unit vector $\mathbf{n} \times \mathbf{c}$. As the conductivity is presumably much less perpendicular to the smectic layer than inside the layer we assume for the third principal axis that it is the unit vector $\mathbf{n}$. Thus the second principal axis is the unit vector $\mathbf{n} \times (\mathbf{n} \times \mathbf{c})$.

(B) Field equations

The equations for the fields are the following:

$$\delta\rho/\delta t = -\operatorname{div} \mathbf{J} \quad \text{(continuity equation for the electric current } J\text{)} \tag{3}$$

here ρ is the electric charge density

$$\rho = \operatorname{div} \mathbf{D} \quad \text{(the second Maxwell equation)} \tag{4}$$

$$\mathbf{J} = (\boldsymbol{\sigma}) \cdot \mathbf{E} \quad \text{(Ohm's law)} \tag{5}$$

From the third assumption for the electric conductivity tensor $\boldsymbol{\sigma}$ with the notation $\sigma_1 - \sigma_2 = \sigma_*$ we have in the XYZ coordinate system:

$$\sigma = \begin{bmatrix} \sigma_1 - \sigma_0 \sin^2 \varphi & (1/2)\sigma_* \sin 2\varphi & 0 \\ (1/2)\sigma_* \sin 2\varphi & \sigma_2 + \sigma_* \sin^2 \varphi & 0 \\ 0 & 0 & \sigma_3 \end{bmatrix} \tag{6}$$

For the electric induction $\mathbf{D}$ we have

$$\mathbf{D} = \boldsymbol{\varepsilon}\mathbf{E} + \mathbf{P}_0 \tag{7}$$

From the second assumption we get that in the XYZ system:

$$\varepsilon = \begin{bmatrix} \varepsilon_1 \cos^2\varphi + \varepsilon_* \sin^2\varphi & (1/2)\sin 2\varphi(\varepsilon_1 - \varepsilon_*) & -\sin\varphi\varepsilon_a \\ (1/2)\sin 2\varphi\cdot(\varepsilon_1 - \varepsilon_*) & \varepsilon_1\sin^2\varphi + \varepsilon_*\cos^2\varphi & \cos\varphi\varepsilon_a \\ -\sin\varphi\cdot\varepsilon_a & \cos\varphi\cdot\varepsilon_a & \varepsilon_+ \end{bmatrix} \quad (8)$$

Here we used the following notations:

$$\varepsilon_* = \varepsilon_2\cos^2\theta_0 + \varepsilon_3\sin^2\theta_0 \quad (8a)$$

$$\varepsilon_a = (1/2)\sin 2\theta_0(\varepsilon_2 - \varepsilon_3) \quad (8b)$$

$$\varepsilon_+ = \varepsilon_2\sin^2\theta_0 + \varepsilon_3\cos^2\theta_0 \quad (8c)$$

The free energy density f is the sum of the following terms:
f_1 is the free energy due to the variation of the layer distances

$$f_1 = (B/2)\cdot[\delta u/\delta z - (1/2)(\delta u/\delta y)^2]^2 \quad (9a)$$

here B is the elastic constant in case of compression
f_2 describes the curvature of the layers

$$f_2 = (K/2)(\delta^2 u/\delta y^2)^2 \quad (9b)$$

(K is the elastic constant in the case of layer bend)
f_3 contains the terms arising from the director curvature

$$f_3 = (K'/2)\cdot(\delta^2\varphi/\delta z^2)^2 + (K''/2)\cdot(\delta\varphi/\delta y)^2 \quad (9c)$$

here $K' = K_B\sin^2\theta\cdot\cos^2\theta + K_T\sin^4\theta$ and $K'' = K_B\sin^4\theta + K_T\sin^2\theta\cdot\cos^2\theta$. ($K_B$ and K_T are the elastic constants for the director bend and twist respectively and θ is the tilt angle between the director and the layer normal.)
f_4 describes the interaction of the electric field and the material

$$f_4 = \int \mathbf{E}\, d\mathbf{D} \quad (9d)$$

The free energy density f is the sum of these terms

$$f = \sum_i f_i \quad (9)$$

We obtain the average free energy F of the sample by averaging f in the y and z directions, namely:

$$F = (1/\lambda)\int_0^\lambda dy(1/d)\int_0^d f\, dz \quad (10)$$

where $\lambda = 2\pi/q$ is the wavelength of the undulation in the y direction.

(C) Calculation

1. Calculation of E_z. Applying an external electric field

$$\mathbf{E}(t) = \mathbf{E}\cdot e^{i\omega t} \quad (11)$$

in nonpertrubed state ($\delta\varphi = \delta u = 0$) from Equations (3)–(8) and Equation (11) we obtain that

$$E_z = -\pi \frac{i\omega\varepsilon_0 E}{\sigma_3 + i\omega\varepsilon_+} (\mathbf{P}_0 \cdot \mathbf{E})/(P_0 \cdot E) = E \cdot \xi \tag{12}$$

The perturbation of $\delta\varphi$ and δu creates an electric field in the y direction. This E_Y modifies the field E_z. However this is a second order effect, which can be neglected in the calculation of the threshold.

2. *Calculation of E_y.* Up to the first order in φ from Equations (1)–(8) after some algebra one gets that

$$E_y = -\varphi\left[E\frac{\sigma_* + i\omega(\varepsilon_1 - \varepsilon_*)}{\sigma_2 + i\omega\varepsilon_*} - P_0\frac{i\omega}{\sigma_2 + i\omega\varepsilon_*}\right] = -\varphi(Ex - P_0\gamma) \tag{13}$$

3. *Calculation of the free energy F.* From Equations (9) and (10) we have:

$$F_1 = (1/8)[Bu_0^2(\pi/d)^2 + (1/64)u_0^4 q^4] \tag{14}$$

$$F_2 = (1/8)Ku_0^2 q^4 \tag{15}$$

$$F_3 = (1/8)K'\varphi_0^2(\pi/d)^2 + (1/8)K''\varphi_0^2 q^2 \tag{16}$$

and from Equations (12), (13) and Equations (1)–(8) up to the lowest order in φ and $\delta u/\delta y$ one gets that

$$F_4 = -(1/8)[\varphi_0^2(E^2 a + P_0^2 b + EP_0 c) + u_0\varphi_0 q(EP_0 e + E^2 h)] \tag{17}$$

Here we used the following notations:

$$a = \varepsilon_*\chi^2 - \chi(\varepsilon_1 - \varepsilon_*),$$
$$b = \gamma + \gamma^2\varepsilon_*$$
$$c = \gamma(\varepsilon_1 - \varepsilon_*) - 2\chi\gamma\varepsilon_* - \chi$$
$$e = \xi + \xi\gamma(\varepsilon_* - \varepsilon_+)$$
$$h = \xi[(\varepsilon_1 - \varepsilon_*) - \chi(\varepsilon_* - \varepsilon_+)]$$

4. *Calculation of E_{th}.* Minimizing with respect to $\delta\varphi$ and δu we get an expression for u_0^2 as the function of E and q. The threshold E_{th} is obtained from the condition $u_0^2 = 0$. This condition leads to equation:

$$B(\pi/d)^2 + Kq^4 = \frac{(E_{th}^2 hq + EP_0 eq)^2}{K''q^2 + K'(\pi/d)^2 - E^2 a - P_0^2 b - EP_0 c} \tag{18}$$

a. First we calculate the threshold field E_{th} in the high frequency range, where the spontaneous polarization cannot follow the electric field that is the permanent dipole moment can be neglected compared to the induced one ($P_0 \ll E\varepsilon_a$). In the case of our material this is fulfilled at frequencies $f > 7$ Hz.

Introducing the notations:

$$S = [B(\pi/d)^2 + Kq^4)(K''q^2 + K'(\pi/d)^2]$$

and

$$L = a(B(\pi/d)^2 + Kq^4)$$

we obtain from Equation (18) that

$$E_{\mathrm{th}}^2 = \frac{-L + (L^2 + 4h^2q^2S)^{1/2}}{2h^2q^2} \tag{19}$$

Using typical elastic constants[10] ($B = 10^8$ J/m^3, $K = 10^{-10}$ J/m, $K' = 10^{-13}$ J/m, and $K'' = 10^{-12}$ J/m) and considering the results obtained for the dielectric constant of FK4[14] ($\varepsilon_* = 4.5\varepsilon_0$ and $\varepsilon_a = -1\varepsilon_0$, ε_0 denotes the vacuum permeability) we find that $4h^2q^2S \ll L^2$ thus we obtain as an approximation for the threshold field E_{th} that:

$$E_{\mathrm{th}} \sim (S/L)^{1/2} \tag{20}$$

Minimizing E_{th} with respect to the wave number q in this approximation we get that $q_c = 0$ i.e. the wavelength of the undulation is infinite. (We regarded a sample with infinite diameters and with perfect alignment in y direction.) However we believe that taking into consideration the finite length of sample thickness (40 μm and 60 μm in the experiments[11]) or the average diameter of the monodomains in y direction (200–300 μm), the calculated value of the q would be in the order of sample thickness and in the diameter of the monodomains. (In the theoretical investigations of instabilities in nematics it is a usual condition that the wavelength of the fluctuating mode cannot be much larger than the sample thickness.[15])

We note however that the value of the minimum threshold field shows no significant difference should we use the $q_c = 0$ or the much more realistic $q_c = 10^4$ 1/m values. Substituting q_c to the expression of E_{th} in Equation (20) in both cases we get that

$$E_{\mathrm{th}}^{\mathrm{min}} = (K'/a)^{1/2}(\pi/d) \sim 10^5 \text{ V/m} \tag{21}$$

The measured values of the threshold of the undulation was 5–10 V for the sample with thickness 60 μm,[11] hence the calculated $E_{\mathrm{th}}^{\mathrm{min}}$ agrees in a good approximation with the experimental value.

b. In the low frequency range ($f < 7$ Hz) $P_0 \gg E\varepsilon_a$ ($P_0 = 10^{-5}$ Q/m^2 at room temperature[14]), taking into consideration the parameters listed in part a., one obtains from Equation (18) that

$$E_{\mathrm{th}} = (K''q^2)/(P_0 e) \tag{22}$$

In this approximation minimizing E_{th} with respect to q, as in the high frequency range one obtains again zero for q_c. However similarly to the paragraph a. with the most realistic approximation, which takes into consideration the sample thickness and the degree of alignment we would get that the wavelength $1/q_c$ is in the order of the diameter of the smectic monodomains. Writing such q_c to Equation (22) we get that $E_{\mathrm{th}}^{\mathrm{min}} \sim 10^4$–$10^5$ V/m. This value again is in a good correspondence with the measured one. (It is in accordance with our observation where we found that the dechiralization lines shift in the z direction except at the walls where domains with slightly different layer normals join together. At these walls the lines are pinned down yielding a bended stripe structure.[11])

We note that N. A. Clark suggested such instabilities even for this geometry[10] and predicted for the threshold field, E_{th}^{min} that $E_{th}^{min} \sim (K''/P_0) \cdot (\pi/d)^2$. Our expression with $q = \pi/d$ corresponds to this one, but we calculated the proportionality coefficient as well (see Equation 22).

III. EXPLANATION OF THE MINIMUM OBSERVED[11] IN THE FREQUENCY DEPENDENCE OF THE UNWINDING CRITICAL FIELD E_c

At fields near to the threshold E_{th}^{min} the bend distortion $\delta\varphi(y, z)$ and layer displacement $\delta u(y, z)$ are sinusoidally varying. Following Clark's arguments[10] we assume that as E increases φ_0 approaches $\pi/2$ and the bend distortion saturates. At sufficiently high fields, the distortion $\delta\varphi(y, 0)$ can be represented by a series of charged π-disclination walls of width w. The wall width $w \ll 1/q_c$ is determined by the balance of $D \cdot E$ and the Frank elastic torques. The deformation of azimuth angle $\delta\varphi(y, z)$ in this limit can be approximated as

$$\delta\varphi(y, z) = (\pi/2)\exp(-(y - 2\pi n/q)^2/w^2)\cos(\pi z/d) \tag{23}$$

Here n is the number of walls counted from $y = 0$.

Consequently, the layer distortion has a zig-zag shape which is due to the body force exerted by the E_z field on the space charge $\rho(y, z)$. This force is confined to regions of width of $\Delta y < w$.

At the electric field E_c the zig-zag structure may disappear due to the ferroelectric or dielectric interactions[16] or as a result of this body force. Generally, in the case of pure samples at low frequencies the ferroelectric coupling, while at high frequencies the dielectric coupling causes the disappearences of the dechiralization lines possibly by the mechanism proposed by Hudak.[17] However our latest measurement on impure samples ($\sigma > 10^{-9}\ 1/\Omega m$) showed that at frequencies near to f_{RC} the unwinding takes place owing to a flow in the z direction.[11] This fact suggests that at these frequencies the unwinding is caused by the above mentioned body force acting on the space charge which appeared due to the conductivity anisotropy. At threshold value F_{th} this force tears the lines. This threshold is determined only by the elastic constants and the degree of deformation, so we can suppose that it does not depend on the frequency.

Thus the basic equation of the unwinding reads:

$$\langle \rho^{th} E_z^{th} \rangle = F_{th}(= \text{independent of frequency}) \tag{24}$$

here $\langle\ \rangle$ denotes time averaging.

If the force $\langle \rho E_z \rangle$ reaches F_{th} at a smaller E_c than required for the ferroelectric or dielectric unwinding mechanism the unwinding takes place by means of the observed flow. With the help of Equations (5) and (3) we can calculate the charge density $\rho(y, z)$. This reads:

$$\rho(y, z) = (1/\omega)\cos(\pi z/d)(\sigma_2(\delta E_y/\delta y) + \sigma_*[E(\delta\varphi/2\delta y)\cos 2\varphi + E_y(\delta\varphi/\delta y)\sin 2\varphi + (\delta E_y/\delta y)\sin^2\varphi]) \tag{25}$$

The unwinding starts at places where the $\rho(y, z)$ function has a maximum, ρ (it is at $z = 0$ and near to $y = w/2$). At these places one obtains for $\langle\rho E_z\rangle$ that:

$$\langle\rho E_z\rangle = \frac{\pi}{2w}E^2\left(\frac{\varepsilon_a\varepsilon + \omega}{\sigma_3^2 + \omega^2\varepsilon_+^2}\left[\sigma_* + \frac{\sigma_*\sigma_2 + \omega^2(\varepsilon_1 - \varepsilon_*)\varepsilon_*}{\sigma_2^2 + \omega^2\varepsilon_*^2}(\sigma_2 + \sigma_*)\right] + \frac{2\varepsilon_a\sigma_3\omega}{\sigma_3^2 + \omega^2\varepsilon_+}\left[(\sigma_2 + \sigma_*)\frac{\sigma_2(\varepsilon_1 - \varepsilon_*) - \sigma_*\varepsilon_*}{\sigma_2 + \omega^2\varepsilon_*^2}\right]\right) \tag{26}$$

Substituting typical constants to Equation (26) we find that the body force $\langle\rho E_z\rangle$ has a maximum at $\omega/2\pi \sim \sigma_3/2\pi\varepsilon_+$, which is practically equal to f_{RC}.

This means that the unwinding critical field $E_c(\omega)$ (which is determined from Equation (24)) shows a minimum at f_{RC}, provided this value is smaller than the unwinding critical field determined from the ferroelectric or dielectric interaction (see Figure 2). The unwinding near to f_{RC} is caused by flow as it was schematically illustrated in Figure 4 of Reference 11. Because in the dielectric regime E_c is much greater than in the ferroelectric one, the unwinding by flow will be effective probably in the dielectric regime as it was observed actually in our experiment.[11] As f_{RC} is proportional to the average conductivity σ, σ must be high enough to ensure that f_{RC} falls into the dielectric regime. E.g. in Reference 11 for the threshold conductivity $\sigma < 10^{-8}\ 1/\Omega\text{m}$ was found.

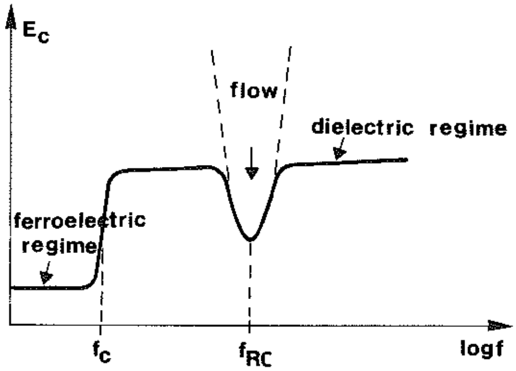

FIGURE 2 The calculated frequency dependence of the unwinding critical field E_c. At low frequencies the spontaneous polarization can follow the electric field, thus the unwinding is caused mainly by the ferroelectric coupling. At high frequencies ($f \gg f_c$) the spontaneous polarization cannot follow the electric field, thus the unwinding is due to only the dielectric coupling. In case of large conductivity, in the vicinity of $f_{RC} = \sigma/(2\pi\varepsilon)$ the unwinding is forced by a flow in the direction of the smectic layer normal n. This calculated frequency dependence is in accordance with the experimental data presented in Reference 11.

IV. SUMMARY

By the simplified theory presented in the paper we could explain the existence of the undulation instability found by us in thick samples of planar oriented S_c^* liquid crystals. On the basis of this simple theory we were able to derive the order of magnitude of the threshold field and to explain the minimum in the frequency dependence of the unwinding critical field E_c as well.

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