Hydrodynamic instabilities of nematic liquid crystals under A. C. electric fields

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HYDRODYNAMIC INSTABILITIES OF NEMATIC LIQUID CRYSTALS UNDER A. C. ELECTRIC FIELDS (*)

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Résumé. — Nous présentons une extension au cas de champs électriques alternatifs de la théorie des instabilités hydrodynamiques due à Helfrich. Les effets électrohydrodynamiques sont décrits par deux équations couplées pour la densité de charge (q) et la courbure locale de l'alignement moléculaire (ψ). Le temps de relaxation pour q est le temps de relaxation diélectrique τ ($\sim 10^{-2}$ s dans les échantillons typiques). Pourvu que l'épaisseur de l'échantillon soit supérieure à une certaine limite d_c, nous trouvons deux domaines de fréquence où la nature de l'instabilité est très différente ; ces domaines sont caractérisés par le rapport de la fréquence du champ électrique, ω, à une fréquence critique, ω_c. Si $\omega < \omega_c$, au seuil d'instabilité, la charge q oscille à la fréquence ω, tandis que la courbure ψ ne dépend pas du temps. Si $\omega > \omega_c$, au seuil d'instabilité, q reste indépendant du temps et ψ oscille. Ces prédictions, et les courbes théoriques de tension-seuil en fonction de ω et d, sont en accord raisonnable avec nombre d'expériences récentes.

Abstract. — We present an extension of the Helfrich theory of hydrodynamic instabilities to the case of alternating electric fields. The electrohydrodynamic effects are described by two coupled equations for the charge density (q) and the local curvature of the molecular alignment (ψ). The relaxation time for q is the dielectric relaxation time τ ($\sim 10^{-2}$ s in typical samples). The relaxation time T for ψ is strongly dependent on the field magnitude. Provided that the sample thickness d is above a certain limit d_c, the nature of the instability is very different, depending on the ratio of the field frequency ω to a critical frequency ω_c. For $\omega < \omega_c$ the onset of instability corresponds to a charge q which oscillates at the frequency ω, while the curvature ψ is essentially time independent. For $\omega > \omega_c$ the situation at threshold corresponds to a constant q and an oscillating ψ. These predictions, together with the calculated curves of threshold voltage vs ω and d, are in reasonable agreement with a number of recent experiments.

I. Introduction. — The effects of electric fields on nematic liquid crystals are spectacular : when a nematic sample with negative dielectric anisotropy

$$\varepsilon_{\parallel} - \varepsilon_{\perp} = \varepsilon_a < 0$$

is placed between semi-transparent electrodes and a voltage V is applied, one observes the following sequence of events (1 − 4) (in increasing V'_s) :

a) At a certain threshold V_c a cellular type of hydrodynamic motion appears : this is revealed by small concomitant deflexions of the molecular alignment, or by the motion of dust particles floating in the liquid.

b) At a higher threshold V_t the flow becomes tur-

bulent ; the molecular alignment is then strongly perturbed, and the scattering of visible light becomes very large : this type of effect has been called « dynamic scattering » by it's discoverers.

In the present paper, we describe some theoretical calculations of the first threshold V_c : this threshold is not the most interesting quantity from the point of view of optoelectronic applications, but it is the best probe available for fundamental studies. Many effects contribute to V_c. A very lucid description of the state of affairs in given in the work of Rondelez [5]. Following his discussion, we must carefully distinguish the cases of DC and AC excitation :

1. FOR DC EXCITATION. — The injection of charges via the electrodes plays an important role. This was already apparent in the work of the RCA group on V_t. More recently, Koelmans and Van Boxtel [6] have shown that there remains a threshold V_{ci} (very similar in magnitude to V_c) in the *isotropic* phase of the nematic : in this phase we are probably

dealing with the « Felici instability » of all liquid insulators under unipolar charge injection [7]. However, in the *nematic* phase, we must have some other processes superimposed to charge injection, since dynamic scattering is observed only for materials with $\varepsilon_a < 0$. The present interpretation of these processes is based on an idea of Carr [8] which has been worked out in great detail by Helfrich [9] ; the principle is explained on figure 1. A qualitative discussion incor-
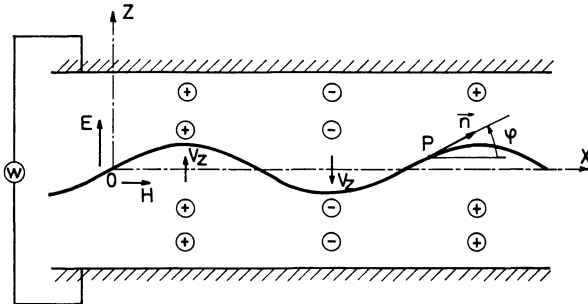

FIG. 1. — A nematic liquid crystal is, at zero electric field, aligned along X-direction. A magnetic field H and an AC external electric field $E(t)$ are respectively applied along Y and Z-direction. d is the sample thickness and φ the fluctuation angle of the director (preferred direction) around X-direction.

porating simultaneously the Felici effect and the Carr-Helfrich effect has been given recently [10].

2. AC VOLTAGES. — At first sight this situation might appear more complex : in actual fact it is simpler, for the following reason : as soon as the frequency $\omega/2\pi$ exceeds a few cycles, charge injection becomes negligible [5]. Thus, by going to AC voltages, we eliminate all the complications related to the nature of the electrodes and the chemical reactions at their contact.

The present paper does not take into account any injection effects : it is thus restricted to the AC regimes. The only new contribution which we bring in, as compared to the work of Helfrich [9], is to introduce time dependent equations, where the time lags associated with the charge response and with the orientational response of the liquid crystal are taken into account. Just as in ref. [9] our description contains certain oversimplifications : if the sample is parallel to the xy plane, with the molecules along x in the unperturbed state and the electric field along z (see Fig. 1), we assume that all physical quantities depend only of x, not of y or z : the only type of deformation which is taken into account is a pure bending mode. This is qualitatively correct, as shown by a number of recent experiments. However, near the walls, the « director » n must also depend on z to satisfy the correct boundary conditions. In our approximate treatment, this is not taken into account ; we simply say, as in ref. [9], that the wavelength of the fluctuating mode along x cannot be much larger

than the sample thickness [1]. A more detailed three dimensional analysis could be attempted, but the present, rough approach allows for much more physical insight.

In section II, we rederive the fundamental dynamic equations describing the charge flow and the changes in molecular orientation, based on the hydrodynamic theories of Ericksen [11] and Leslie [12], [13]. (We consider only materials where the anisotropy of the electrical conductivity $\sigma_a = \sigma_{\parallel} - \sigma_{\perp}$ is positive — as it has always been found up to now.) In section III, we apply these equations to the low frequency regime, where the charges are modulated at the frequency ω of the electric field. We call this the « conducting regime ». In section IV, we discuss the high frequency regime where the charge distribution becomes essentially static (« non conducting regime »). In section V we discuss some extension of our results to systems where the dielectric anisotropy would be slightly positive, rather than negative, but where the unperturbed molecular alignment is still in the plane of the slab (because of the walls or because of a magnetic field H applied along x). The case where the applied electric field, instead of being a sine wave, is a succession of rectangular pulses, will be the object of a second paper : it leads to a very different diagram of threshold versus frequency, and could be of some experimental interest.

II. **Electrohydrodynamic equations.** — As explained in the introduction, we consider a nematic slab of thickness d, lying in the xy plane (Fig. 1) and submitted to an electric field $E = E_M \cos \omega t$ along z. We assume that in the unperturbed state the molecules are aligned along the x direction, and we also allow for a stabilising magnetic field H applied along x. We consider pure bending fluctuations [2] where the director n is in the (x, z) plane, making a small angle φ with the x direction. Finally we assume that φ is only a function of x.

Associated with such an orientational fluctuation, we have a space charge $q(x)$ and a hydrodynamic flow velocity : in our approximation where everything depends only on x, this velocity has only one non-zero component $v_z(x)$ [3].

For the internal rotation, we can neglect the inertial terms. Let us define as in ref. [14] the viscous torque as the frictional torque exerted by the molecules on the over-all hydrodynamic motion. This viscous torque must be equal to the sum of the elastic,

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[1] For some cases, we will find that the wavelength is in fact much smaller than the thickness : then our analysis is probably quite correct.

[2] Helfrich has shown that, for this orientation of the electric field, these pure bending fluctuations are the only ones to induce shear flows.

[3] For the slow motions of interest, the nematic may be considered as incompressible : here this imposes $\partial v_x/\partial x = 0$ and finally $v_x = 0$ when there is no general drift along x.

magnetic and dielectric torques exerted on the molecules :

$$\Gamma_{\text{visc.}} = \Gamma_{\text{el.}} + \Gamma_{\text{magn.}} + \Gamma_{\text{diel.}} .$$

The dielectric torque is induced by the total electric field, i. e. the sum of the external field and the field due to space charges q.

II.1 CONTRIBUTIONS TO THE TORQUE.

— The electric, dielectric and magnetic torques can be derived from the free-energy

$$F = \int_V g(\mathbf{r}) \, dv$$

$$g(\mathbf{r}) = \frac{1}{2} \left[K_{11}(\text{div } \mathbf{n})^2 + K_{22}(\mathbf{n}.\text{curl } \mathbf{n})^2 + \right.$$

$$\left. + K_{33}(\mathbf{n} \times \text{curl } \mathbf{n})^2 - \frac{\varepsilon_a}{4\pi} (\mathbf{n}.\mathbf{E})^2 - \chi_a(\mathbf{n}.\mathbf{H})^2 \right]$$

where K_{ii} are the elastic coefficients in Franck's [15] notation, χ_a and ε_a the anisotropic parts of the magnetic susceptibility and dielectric constants

$$\chi_a = \chi_\parallel - \chi_\perp ; \qquad \varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp .$$

It will be assumed that the fluctuation angle, φ, is small : only first order terms in φ will be kept. This fluctuation induces a space charge q which in turn induces the x-component E_x of the total electric field [9]. As shown by Helfrich, E_x and q are first-order terms in φ. For the sake of simplicity, we will omit the part of the z-component of the electric field induced by the space-charge q : it can be easily shown that this component occurs only in second-order terms in φ. Hence $E_z = E(t)$.

In our frame of reference the torques are given by the functional derivative

$$\Gamma_y(\mathbf{r}) = \frac{\delta F}{\delta \varphi(\mathbf{r})} .$$

Assuming a periodic fluctuation

$$\varphi = \varphi_0 \cos kx$$

one finds, for the elastic, magnetic and dielectric torques :

$$\Gamma_{\text{el.},y} = -K_{33} \frac{\partial^2 \varphi}{\partial x^2} = K_{33} \, k^2 \, \varphi$$

$$\Gamma_{\text{magn.},y} = \chi_a \, H^2 \, \varphi \qquad\qquad (\text{II, 2})$$

$$\Gamma_{\text{diel.},y} = -\frac{\varepsilon_a}{4\pi} E^2 \left(\varphi + \frac{E_x}{E} \right).$$

Using the notations of reference [14], the viscous torque is

$$\Gamma_{\text{visc.}} = \mathbf{n} \times (\gamma_1 \, \mathbf{N} + \gamma_2 \, \overline{\overline{A}}.\mathbf{n})$$

with

$$\mathbf{N} = \left(\mathbf{\Omega} - \frac{1}{2} \text{curl } \mathbf{v} \right) \times \mathbf{n}$$

$$A_{ij} = \frac{1}{2} \left[\frac{\partial v_i}{\partial r_j} + \frac{\partial v_j}{\partial r_i} \right]$$

where the director $\mathbf{n}(\mathbf{r})$ is a unit vector along the preferred orientation of the molecule ; $\mathbf{\Omega}$ is the angular velocity of the director and $\mathbf{v}$ is the fluid velocity. Recalling that the only non-zero component of $\mathbf{v}$ is $v_z(x, t)$ the only non-zero components of tensor $\overline{\overline{A}}$ are

$$A_{zx} = A_{xz} = \frac{1}{2} \frac{\partial v_z}{\partial x} .$$

Hence

$$\Gamma_{\text{visc.},y} = -\gamma_1 \left[\dot{\varphi} - \frac{\gamma_1 - \gamma_2}{2\gamma_1} \frac{\partial v_z}{\partial x} \right]. \qquad (\text{II, 3})$$

We will use further the following notations

$$D_0^2 = -\varepsilon_a \, E_0^2 = 4\pi \frac{\varepsilon_\parallel}{\varepsilon_\perp} [\chi_a \, H^2 + K_{33} \, k^2] . \qquad (\text{II, 4})$$

The minus sign in the definition of E_0^2 will be useful in sections III and IV where we assume a negative dielectric anisotropy, which is the case for the experimental data. In section V, we will allow ε_a to be positive, and use only D_0^2.

Inserting eqs. (II, 2-4) into eq. (II, 1), one finds

$$-\Gamma_{\text{visc.},y} = \gamma_1 \left[\dot{\varphi} - \frac{\gamma_1 - \gamma_2}{2\gamma_1} \frac{\partial v_z}{\partial x} \right] =$$

$$= \frac{\varepsilon_a}{4\pi} \left[\left(\frac{\varepsilon_\perp}{\varepsilon_\parallel} E_0^2 + E^2 \right) \varphi + EE_x \right]. \qquad (\text{II, 5})$$

II.2 CHARGE-BALANCE.

— The conservation equation for charges is

$$\dot{q} + \frac{\partial J_x}{\partial x} = 0 \qquad\qquad (\text{II, 6})$$

where q is the excess charge per unit volume and $\mathbf{J}$ the electric current. It is shown in Appendix A that the diffusion currents can be neglected. Then

$$\mathbf{J} = \sigma_\perp \mathbf{E} + \sigma_a \, \mathbf{n}(\mathbf{n}.\mathbf{E}) . \qquad (\text{II, 7})$$

Keeping only first-order terms in φ and E_x,

$$J_x = \sigma_\parallel E_x + \sigma_a E\varphi . \qquad (\text{II, 8})$$

From the relation

$$\frac{\partial D_x}{\partial x} = \frac{\partial}{\partial x} (\varepsilon_\parallel E_x + \varepsilon_a E\varphi) = 4\pi q \qquad (\text{II, 9})$$

one obtains

$$\frac{\partial E_x}{\partial x} = \frac{4\pi q}{\varepsilon_\parallel} - \frac{\varepsilon_a}{\varepsilon_\parallel} E\psi \qquad (\text{II, 10})$$

where $\psi = \partial\varphi/\partial x$ is the local curvature. Inserting eqs. (II, 8) and (II, 10) into eq. (II, 6), one has the charge-balance equation

$$\dot{q} + \frac{q}{\tau} + \sigma_H E\psi = 0 \qquad (\text{II, 11})$$

with

$$\begin{cases} \dfrac{1}{\tau} = \dfrac{4\,\pi\sigma_\parallel}{\varepsilon_\parallel} \\[2mm] \sigma_H = \sigma_\parallel \left(\dfrac{\varepsilon_\perp}{\varepsilon_\parallel} - \dfrac{\sigma_\perp}{\sigma_\parallel} \right). \end{cases} \qquad \text{(II, 12)}$$

Here τ is the dielectric relaxation time for charges. Eq. (II, 11) relates the charge q to the curvature ψ.

II.3 ACCELERATION EQUATION. — The equation of motion is [4]

$$\rho \frac{\partial \mathbf{v}}{\partial t} = - \operatorname{Div}(\rho \mathbf{vv}) + \operatorname{Div}(\bar{\bar\sigma} + \bar{\bar\sigma}') + q\mathbf{E} \qquad \text{(II, 13)}$$

where $\bar{\bar\sigma}$ and $\bar{\bar\sigma}'$ are the elastic and viscous stress tensor [11], [15]

$$\begin{cases} \sigma_{ij} = -p\delta_{ij} - \sum_l \left(\dfrac{\partial g_{\text{el.}}}{\partial n_{l,i}} \right) n_{l,j} \\[2mm] g_{\text{el}} = \dfrac{1}{2}[K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}.\operatorname{curl}\mathbf{n})^2 + \\ \qquad\qquad + K_{33}(\mathbf{n}\times\operatorname{curl}\mathbf{n})^2] \quad \text{(II, 14)} \\[2mm] \bar{\bar\sigma}' = \alpha_1(\mathbf{n}.\bar{\bar A}.\mathbf{n})\,\mathbf{nn} + \dfrac{\gamma_2 - \gamma_1}{2}\mathbf{nN} + \dfrac{\gamma_2 + \gamma_1}{2}\mathbf{Nn} + \\[2mm] \qquad + \alpha_4\bar{\bar A} + \dfrac{\beta - \gamma_2}{2}\mathbf{n}(\mathbf{n}.\bar{\bar A}) + \dfrac{\beta + \gamma_2}{2}(\bar{\bar A}.\mathbf{n})\,\mathbf{n}. \end{cases}$$

We use for $\bar{\bar\sigma}'$ the notation of ref. [16] : this takes into account the effect of the Onsager reciprocal relations on the expression of the viscous stress tensor. $\mathbf{N}$ and $\bar{\bar A}$ are defined in section (II, 1).

The x-component of eq. (II, 13) defines the pressure p. We are interested only in the z-component of this equation. Since $v_x = 0$ and $n_{1,z} = 0$,

$$(\operatorname{Div}\rho\,\mathbf{vv})_z = \frac{\partial}{\partial x}(\rho v_x\,v_z) = 0$$

$$(\operatorname{Div}\bar{\bar\sigma})_z = \frac{\partial}{\partial x}\sigma_{xz} = \frac{\partial}{\partial x}\sum_l \left(\frac{\partial g_{\text{el.}}}{\partial n_{l,x}} \right) n_{l,z} = 0.$$

Hence

$$\rho \frac{\partial v_z}{\partial t} = \frac{\partial}{\partial x}\sigma'_{xz} + qE. \qquad \text{(II, 15)}$$

Keeping only first-order terms in eq. (II, 14), σ'_{xz} is given by

$$\sigma'_{xz} = +\left(\frac{\gamma_1 - \gamma_2}{2\,\gamma_1} \right) \Gamma_{\text{visc.}} + \eta' \frac{\partial v_z}{\partial x} \qquad \text{(II, 16)}$$

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[4] We use the following notations [11], [16]

— ab is a dyadic with the components $(ab)_{ij} = a_i\,b_j$
— $\mathbf{a}.\bar{\bar T}$ is a vector :

$$(\mathbf{a}.\bar{\bar T})_i = \sum_j a_j\,T_{ji}, \quad (\bar{\bar T}.\mathbf{a})_i = \sum_j T_{ij}\,a_j$$

— f, j is the functional derivative $\delta f/\delta x_j$
— $(\operatorname{Div}\bar{\bar T})$ is a vector with components $(\operatorname{Div}\bar{\bar T})_i = \sum_j T_{ji,j}$.

where

$$\eta' = \frac{\gamma_1(2\,\alpha_4 + \beta) - \gamma_2^2}{4\,\gamma_1}. \qquad \text{(II, 17)}$$

From thermodynamical relations [16], η' is positive definite. Inserting (II, 16) in (II, 15), we find, for the acceleration equation :

$$\rho\dot{v}_z = \frac{\gamma_1 - \gamma_2}{2\,\gamma_1} \frac{\partial}{\partial x}\Gamma_{\text{visc.}} + \eta' \frac{\partial^2 v_z}{\partial x^2} + qE. \qquad \text{(II, 18)}$$

In this equation the left h. s. may be neglected (a) in the « conduction » regime where this term vanishes (b) in the « dielectric » regime where $\rho\dot{v}_z$ is of order $n\omega v_z$ and can be neglected with respect to

$$\eta' \frac{\partial^2 v_z}{\partial x^2}$$

which is of order $\eta'\,k^2\,v_z$ (Typical experimental conditions are $\omega \simeq 10^2 - 10^4\,H_z$; $k \simeq 2 \times 10^4\,\text{cm}^{-1}$; $\eta' \simeq 10^{-1}$ poise).

II.4 EQUATION FOR THE CURVATURE. — Using eqs. (II, 2), (II, 3), (II, 10) and (II, 18), the x-derivatives of the torques can now be expressed as functions of the charge q and the curvature ψ. Let us introduce the following notations :

$$\begin{cases} \eta_0 = \left(\dfrac{2\,\gamma_1}{\gamma_1 - \gamma_2} \right)^2 \eta' \\[2mm] w = -\dfrac{\gamma_1 - \gamma_2}{2\,\gamma_1} \dfrac{\partial^2 v}{\partial x^2} \\[2mm] \lambda = -\dfrac{\varepsilon_a}{4\,\pi} \dfrac{\varepsilon_\perp}{\varepsilon_\parallel} \left(\dfrac{1}{\gamma_1} + \dfrac{1}{\eta_0} \right) \end{cases} \qquad \text{(II, 19)}$$

$$\Gamma_{\text{tot.}} = \Gamma_{\text{el.}} + \Gamma_{\text{magn.}} + \Gamma_{\text{diel.}} - \Gamma_{\text{visc.}}.$$

Using eqs. (II, 3) and (II, 18) one finds

$$w = -\frac{\gamma_1}{\gamma_1 + \eta_0}\dot{\psi} + \frac{2\,\gamma_1}{(\gamma_1 - \gamma_2)(\gamma_1 + \eta_0)}qE \qquad \text{(II, 20)}$$

$$-\frac{\partial \Gamma_{\text{visc.}}}{\partial x} = \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\left[\dot{\psi} + \frac{1}{\eta_0}\frac{2\,\gamma_1}{\gamma_1 - \gamma_2}qE \right]. \qquad \text{(II, 21)}$$

Using eqs. (II, 2), (II, 4) and (II, 10), one has

$$\frac{\partial \Gamma_{\text{diel.}}}{\partial x} =$$

$$= \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\left[\lambda E^2\,\psi - \frac{\varepsilon_a}{\varepsilon_\parallel}\left(\frac{1}{\gamma_1} + \frac{1}{\eta_0} \right)Eq \right] \qquad \text{(II, 22)}$$

$$\frac{\partial \Gamma_{\text{magn.}}}{\partial x} + \frac{\partial \Gamma_{\text{el.}}}{\partial x} = \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\lambda E_0^2. \qquad \text{(II, 23)}$$

Hence, from eq. (II, 1) one finds the curvature equation

$$\frac{\partial \Gamma_{\text{tot.}}}{\partial x} =$$

$$= \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\left[\dot{\psi} + \lambda(E^2 + E_0^2)\,\psi + \frac{qE}{\eta} \right] = 0 \qquad \text{(II, 24)}$$

where η defined by :

$$\frac{1}{\eta} = \frac{1}{\eta_0} \left(\frac{2\,\gamma_1}{\gamma_1 - \gamma_2} \right) - \frac{\varepsilon_a}{\varepsilon_\parallel} \left(\frac{1}{\gamma_1} + \frac{1}{\eta_0} \right) \qquad (II, 25)$$

is an effective viscosity. From theoretical considerations on flow-alignment of nematic liquid crystals, one can deduce the following inequalities [13]

$$\gamma_1 - \gamma_2 > 2\,\gamma_1 > 0 \,; \quad 0 < - (\gamma_1 + \gamma_2) \ll 1 \,.$$

Hence $2\,\gamma_1/(\gamma_1 - \gamma_2)$ is of order unity. For small values of ε_a $\left(\dfrac{\varepsilon_a}{\varepsilon_\parallel} \ll 1 \right)$,

$$\eta \simeq \eta_0 \simeq \eta' \simeq \frac{2\,\alpha_4 + \beta - \gamma_1}{4} \,.$$

On the contrary, for materials having a strong positive dielectric anisotropy, η can be negative. $1/\eta$ vanishes for

$$\frac{\varepsilon_a}{\varepsilon_\parallel} = \left(\frac{2\,\gamma_1}{\gamma_1 - \gamma_2} \right) \frac{\gamma_1}{\gamma_1 + \eta_0} \qquad (II, 26)$$

i. e.

$$\frac{\varepsilon_\parallel}{\varepsilon_a} \simeq 1 + \frac{\gamma_1}{\eta_0} \,.$$

We shall see in section V the consequences of this point.

Eq. (II, 24) can now be rewritten as :

$$\dot{\psi} + \frac{1}{T}\psi + \frac{qE}{\eta} = 0 \qquad (II, 27)$$

where the time-dependent quantity

$$\frac{1}{T} = \lambda(E^2 + E_0^2) \qquad (II, 28)$$

appears as a relaxation rate for the curvature.

II.5 FORMAL MATHEMATICAL PROPERTIES. — Eq. (II, 11) and (II, 27) are two coupled equations for the charge density q and the curvature ψ. They are linear in q and ψ ; from a mathematical point of view, they have some similarities with the spinor equations describing the motion of a quantum mechanical spin $S = \frac{1}{2}$ in a radiofrequency field. However, one further complication is present : the relaxation rate $1/T$ for the curvature is not a constant, because E is modulated in time. Let assume a sinusoidal field $E = E_M \cos \omega t$ and rewrite our two equations for this case

$$\dot{q} + \frac{q(t)}{\tau} + \sigma_H E_M \psi(t) \cos \omega t = 0 \qquad (II, 29)$$

$$\dot{\psi} + \lambda(E_0^2 + E_M^2 \cos^2 \omega t) \psi(t) +$$

$$+ \frac{E_M}{\eta} q(t) \cos \omega t = 0 \,. \qquad (II, 30)$$

We shall also find the following notations useful

$$\left\{ \begin{array}{l} \dfrac{1}{T_f} = \dfrac{\lambda E_M^2}{2} = \overline{\lambda E^2(t)} \\[2mm] \mu^2 = \dfrac{E_0^2}{E_M^2} \,. \end{array} \right. \qquad (II, 31)$$

The system (II, 29), (II, 30) accepts solutions of the form

$$\begin{bmatrix} q(t) \\ \psi(t) \end{bmatrix} = \begin{bmatrix} a(t) \\ b(t) \end{bmatrix} e^{st} \qquad (II, 32)$$

where $a(t)$ and $b(t)$ are periodic functions of time with period $2\,\pi/\omega$, and s is a number which may in principle, be real or complex. The proof of eq. (II, 31) is similar to the proof of the Bloch-Floquet theorem for electron propagation in one dimensional solids and is given in appendix B. For each value of E_M, there are two independent solutions of the form (II, 32) with different parameters $s_1(E_M)$ and $s_2(E_M)$ — (We choose to call s_1 the parameter with the largest real part). We are interested here in the onset of instability. This occurs when

$$\mathrm{Re}\,(s_1(E_M)) = 0 \qquad (II, 33)$$

eq. (II, 33) (where Re means : real part of) is an implicit equation for the threshold field E_M.

We shall now proceed to a more quantitative (and less formal) discussion of the threshold in some simple regimes. For these regimes we find that, at threshold, the imaginary part of s_1 is equal to zero : the onset of instability is associated with very slow hydrodynamic motions. This property seems to be in agreement with the experimental observations [5], but we have not been able to prove it for all regimes.

III. Low frequencies and low fields : « conduction regime ». — III.1 DEPENDENCE OF THE CURVATURE $\psi(t)$ ON TIME. — In the present section we shall be concerned with frequencies ω comparable to the dielectric relaxation rate $1/\tau$. It will turn out that, for such frequencies, the electric field threshold is rather small ; then the relaxation time T_f for the molecular orientation (as defined by II, 31) is long

$$\left. \begin{array}{l} T_f \gg \tau \\ \omega T_f \gg 1 \end{array} \right\} \,. \qquad (III, 1)$$

We shall now prove that when the inequalities (III, 1) are satisfied, the curvature ψ (at threshold) is essentially independent of time. Our starting point is eq. (II, 30) which may be rewritten as

$$T_f \dot{\psi} + (2\,\mu^2 + 1 + \cos 2\,\omega t) \psi(t) =$$

$$= - \frac{2}{\lambda\eta E_M} q(t) \cos \omega t \,. \qquad (III, 2)$$

This may be transformed into the integral form

$$\psi(t) = - \frac{2}{\lambda \eta E_M T_f} \exp\left[- (1 + 2\,\mu^2)\frac{t}{T_f} - \frac{\sin 2\,\omega t}{2\,\omega T_f} \right]$$

$$\times \int_{-\infty}^{t} dt'q(t')\cos \omega t'$$

$$\times \exp\left[(1 + 2\,\mu^2)\frac{t'}{T_f} + \frac{\sin 2\,\omega t'}{2\,\omega T_f} \right]$$

(III, 3)

the lower limit of the integral ensures $\psi(t)$ to be a periodic function of t. In the low-field limit $(\omega T_f \gg 1)$, we can make two approximations :

i) $\dfrac{\sin 2\,\omega t}{2\,\omega T_f}$ is of order $\dfrac{1}{2\,\omega T_f}$. We can then take

$$\exp \frac{\sin 2\,\omega t}{2\,\omega T_f} = 1$$

in eq. (III, 3).

ii) The term $\exp(1 + 2\,\mu^2)\, t/T_f$ can be taken as a constant over one period. This means that $q(t) \cos \omega t$ can be replaced in the integral by its average value over one period.

Let us expand $q(t)$ as a Fourier series :

$$q(t) = \sum_{n=0}^{\infty} q'_n \cos n\omega t + q''_n \sin n\omega t .$$

(III, 4)

Hence

$$< q(t) \cos \omega t >_{\mathrm{per.}} = \tfrac{1}{2} q'_1 .$$

After integration one obtains

$$\psi = - \frac{q'_1}{\lambda \eta E_M(1 + 2\,\mu^2)} .$$

(III, 5)

Thus, in this regime, the curvature ψ is not time-dependent : the molecular alignment has a long response time T_f and does not follow the time variation of the field $E(t)$.

III.2 MODULATION OF THE SPACE CHARGE AND THRESHOLD CONDITION.

— Knowing that ψ is independent of t, eq. (II, 29) is easily integrated and gives

$$q(t) = - \frac{\sigma_H E_M \psi \tau}{1 + \omega^2\,\tau^2} (\cos \omega t + \omega \tau \sin \omega t) .$$

(III, 6)

The space charge q oscillates at the frequency ω, but the local force density qE has a DC component $q'_1 E_M/2$. The onset of instability is thus caracterized by a charge oscillation. This regime is then legitimately called a « conduction regime ».

The instability threshold is obtained from eqs. (III, 6) and (III, 5). From eq. (III, 6),

$$q'_1 = - \frac{\sigma_H E_M \tau}{1 + \omega^2\,\tau^2}\,\psi .$$

eq. (III, 5) then gives the threshold condition :

$$1 + 2\,\mu^2 = \frac{\zeta^2}{1 + \omega^2\,\tau^2}$$

(III, 7)

where

$$\zeta^2 = \frac{\sigma_H \tau}{\lambda \eta} .$$

(III, 8)

Inserting eq. (II, 31) into eq. (III, 7), one finds for the threshold field

$$\overline{E^2(t)} = \frac{E_M^2}{2} = \frac{E_0^2(1 + \omega^2\,\tau^2)}{\zeta^2 - (1 + \omega^2\,\tau^2)} .$$

(III, 9)

III.3 DISCUSSION.

— The threshold field depends only on two parameters which caracterize the nematic material. The first one is the relaxation time for charges, τ. The second one is the dimensionless coefficient ζ^2.

$$\zeta^2 = \left[1 - \frac{\sigma_\perp}{\sigma_\parallel} \frac{\varepsilon_\parallel}{\varepsilon_\perp} \right] \left[1 - \frac{\varepsilon_\parallel}{\varepsilon_a} \frac{2\,\gamma_1^2}{(\gamma_1 - \gamma_2)(\gamma_1 + \eta_0)} \right]$$

(III, 10)

(For M B B A $(\sigma_\parallel/\sigma_\perp = 1,5$; $\varepsilon_\parallel = 4,7$; $\varepsilon_\perp = 5,4$; $\eta_0/\gamma_1 \simeq 0,4)$ one finds $\zeta^2 \simeq 3,2$.) The role of the parameter

$$\theta_H = \varepsilon_a(\zeta^2 - 1)$$

has been pointed out by Helfrich [9] for the DC regime. For this reason we shall refer to θ_H as the *Helfrich parameter*. It will play a special role in the discussion on the instabilities for negative or positive ε_a which is given in section V.

It is interesting to note that (in spite of the underlying approximation $\omega T_f \gg 1$) eq. (III, 9) gives the exact threshold in the limit $\omega \to 0$. This DC threshold can be directly obtained from eq. (II, 29-30) :

$$E_{DC}^2 = \frac{E_0^2}{(\zeta^2 - 1)} .$$

(III, 11)

For increasing ω, eq. (III, 9) leads to a « cut-off frequency »

$$\omega_c = \frac{(\zeta^2 - 1)^{1/2}}{\tau} .$$

(III, 12)

For ω increasing toward ω_c, the threshold field tends to become very large : in fact eq. (III, 9) becomes incorrect when E_M reaches a value E_{Mc} such as $1/T_f$ becomes comparable to ω. On figure 2 is shown the range of validity of eq. (III, 9). The approximation will be valid up to $\omega = \omega'_c$. ω'_c will be close to ω_c if $\lambda E_0^2/\omega_c \ll 1$. Let us discuss this quantitatively for $H = 0$ and assuming values of $(\zeta^2 - 1)$ of order unity. Then, if d is the sample thickness, replacing (tentatively) the wave-vector k by π/d as in reference [9], we have

$$\frac{\lambda E_0^2}{\omega_c} \simeq \frac{\pi^2}{d^2} \frac{K_{33}}{\eta''} \tau \ll 1$$

where

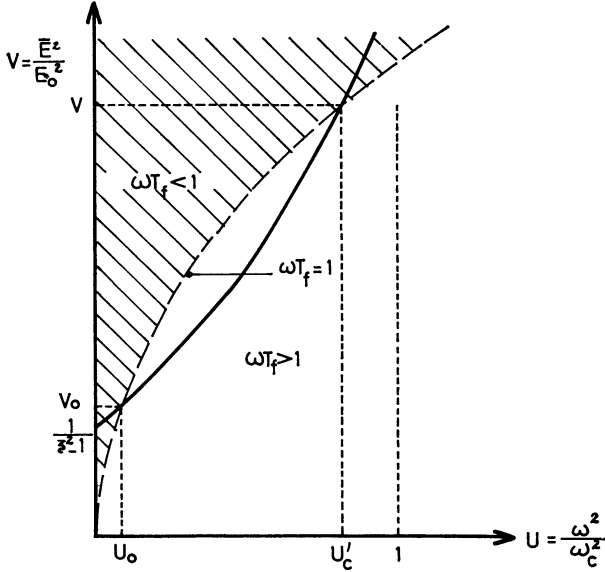$$\frac{1}{\eta''} = \frac{1}{\gamma_1} + \frac{1}{\eta_0} .$$

Fig. 2. — The full line gives the threshold-field of instability versus ω^2 in reduced coordinates. The dashed line is the parabola $u = (\zeta^2 - 1)$ $(d_c/d)^4 v^2$ corresponding to the condition $\omega T_f = 1$. The two lines intersect at points (u_0, v_0) and (u'_c, v_c). The assumption $\omega T_f > 1$ is valid in the corresponding frequency range (ω_0, ω'_c).

Thus one condition for the existence of the low frequency, conducting regime is

$$d \gg d_c$$

where $d_c = \pi(K_{33} \tau/\eta'')^{1/2}$. Typically, $K_{33} \simeq 10^{-6}$ dynes, $1/\tau \simeq 2 \times 10^2$ s^{-1}, $\eta'' = 10^{-1}$ poise ; this gives $d_c \simeq 7 \mu$...

It must be noted that, as can be seen on figure 2, this condition ensures the validity of eq. (III, 9) not only up to frequency $\omega = \omega_c$ but also in the low frequency range. For $d/d_c \ll 1$, one finds for the limiting frequencies

$$\omega_0 = \frac{\omega_c}{(\zeta^2 - 1)^{1/2}} \left(\frac{d_c}{d}\right)^2 ;$$

$$E_{MO}^2 = \frac{E_0^2}{\zeta^2 - 1} \left[1 + \frac{\zeta^2}{(\zeta^2 - 1)^3} \left(\frac{d_c}{d}\right)^4\right]$$

$$\omega'_c = \omega_c \left[1 - \frac{\zeta^2}{2(\zeta^2 - 1)^{3/2}} \left(\frac{d_c}{d}\right)^2\right] ;$$

$$E_{Mc}^2 = E_0^2 (\zeta^2 - 1)^{1/2} \left(\frac{d}{d_c}\right)^2 .$$

Taking $\zeta^2 = 3$; $1/\tau = 200$ s^{-1} ; $d = 100 \mu$; $d_c = 7 \mu$, one finds $\omega_0/2\pi \simeq 0,1$ Hz ; $\omega_c/2\pi \simeq 50$ Hz ;

$$(\omega_c - \omega'_c) \simeq 0,5 \text{ Hz} .$$

Thus, for materials having a negative dielectric anisotropy, the « conduction regime » occurs in the frequency range $(0, \omega_c)$. (For very low frequencies further complications might be caused by carrier injection, but we do not consider this here.)

We shall find in section IV that, for frequencies $\omega > \omega_c$, another regime occurs, the so-called « dielectric » or « fast turn-off » regime. ω_c thus appears as a limiting frequency between two regimes rather than a real cut-off frequency.

We shall now discuss briefly the dependence of the threshold on thickness and on magnetic field. The equivalent field E_0 is given by eq. (II, 4)

$$E_0^2 = -\frac{4\pi}{\varepsilon_a} \frac{\varepsilon_{||}}{\varepsilon_\perp} (\chi_a H^2 + K_{33} k^2)$$

where $k = k_x$ is the wave-vector along x-axis. Both experimental conditions and qualitative physical arguments suggest that $k \simeq \pi/d$ where d is the sample thickness. Hence

$$E_0^2 = -\frac{4\pi}{\varepsilon_a} \frac{\varepsilon_{||}}{\varepsilon_\perp} \left(\chi_a H^2 + K_{33} \frac{\pi^2}{d^2}\right)$$

or, introducing the magnetic coherence length

$$\xi = \frac{1}{H} \sqrt{\frac{K_{33}}{\chi_a}} \qquad (III, 13)$$

$$E_0^2 = -\frac{4\pi}{\varepsilon_a} \frac{\varepsilon_{||}}{\varepsilon_\perp} \frac{K_{33} \pi^2}{d^2} \left[1 + \frac{d^2}{\pi^2 \xi^2}\right]. \qquad (III, 14)$$

a) *For low magnetic fields* $(\xi \gg d)$. — From eqs. (III, 9) and (III, 14), one finds, for the rms threshold voltage V_{thr} :

$$V_{thr}^2(\omega) = \frac{V_0^2(1 + \omega^2 \tau^2)}{\zeta^2 - 1 - \omega^2 \tau^2} \qquad (III, 15)$$

where

$$V_0 = \left[-\frac{4\pi}{\varepsilon_a} \frac{\varepsilon_{||}}{\varepsilon_\perp} K_{33} \pi^2\right]^{1/2} . \qquad (III, 16)$$

Eq. (III, 15) shows that, for low magnetic fields, the experiments will give a voltage threshold independent of sample thickness. This is well confirmed by the experimental results [1], [2].

b) *For higher magnetic fields.* — One has

$$V_{thr}(\omega, H) = V_{thr}(\omega, 0) \sqrt{1 + \frac{d^2}{\pi^2 \xi^2}}$$

or

$$V_{thr}(\omega, H) = V_{thr}(\omega, 0) \sqrt{1 + \frac{H^2}{H_0^2}}$$

where

$$H_0 = \frac{\pi}{d} \sqrt{\frac{K_{33}}{\chi_a}}$$

(with $d = 100 \mu$, $H_0 \simeq 10^3$ G).

For $H \gg H_0$, $V_{thr}(\omega, H)$ will be proportional to d ; one will observe, for fixed ω, a *field* threshold proportional to H. These results are in reasonable agreement with the experiments made at Orsay [5].

IV. « **Dielectric regime** » ($\omega\tau \gg 1$). — When $\omega\tau$ is large, the charges accumulated by the Carr-Helfrich process do not have enough time to flow during one cycle : the charge density q (at threshold) becomes time independent. This may be proved explicitely on an integral version of eq. (II, 29) :

$$q = - \sigma_H E_M e^{-t/\tau} \int_{-\infty}^{t} e^{t'/\tau} \psi(t') \cos \omega t' \, dt' . \quad (IV, 1)$$

In the limit $\omega\tau \gg 1$, $\exp(t'/\tau)$ can be taken as a constant over one period. Thus we can replace in the integral $\psi(t') \cos \omega t'$ by its average value over a period. Expanding $\psi(t)$ as a Fourier series

$$\psi(t) = \sum_{n=0}^{\infty} \psi'_n \cos n\omega t + \psi''_n \sin n\omega t$$

one has

$$< \psi(t) \cos \omega t >_{\text{per.}} = \frac{\psi'_1}{2}$$

and

$$q = - \frac{\sigma_H E_M \tau}{2} \psi'_1 . \quad (IV, 2)$$

The space-charge is time independent. This regime is a « non conducting » regime. Using eq. (IV, 2), eq. (II, 30) is easily integrated :

$$\psi(t) = \frac{\zeta^2}{\omega T_f} \psi'_1 \exp\left[- (1 + 2\mu^2)\frac{t}{T_f} - \frac{\sin 2\omega t}{2\omega T_f} \right] \times$$

$$\times \int_{-\infty}^{\omega t} du \cos u \exp\left[(1 + 2\mu^2)\frac{u}{\omega T_f} + \frac{\sin 2u}{2\omega T_f}\right]$$

$$\quad (IV, 3)$$

where

$$\psi'_1 = \frac{\omega}{\pi} \int_0^{2\pi/\omega} \psi(t) \cos \omega t \, dt . \quad (IV, 4)$$

Eq. (IV, 3) and (IV, 4) lead to the compatibility equation

$$\frac{1}{\zeta^2} = g(X, Y) \quad (IV, 5)$$

where

$$X = \frac{1}{\omega T_f} = \frac{\lambda E_M^2}{2\omega} \quad (IV, 6)$$

$$Y = \frac{\mu^2}{\omega T_f} = \frac{\lambda E_0^2}{2\omega} \quad (IV, 7)$$

$$g(X, Y) = \frac{X}{\pi} \int_0^{2\pi} du \cos u$$

$$\times \exp\left[- (X + 2Y)u - \frac{X}{2}\sin 2u \right] \int_{-\infty}^{u} du' \cos u'$$

$$\times \exp\left[(X + 2Y)u' + \frac{X}{2}\sin 2u'\right]. \quad (IV, 8)$$

In eq. (IV, 5), ζ is a caracteristic of the material. E_0^2 and Y depend on the wavevector k. For fixed ω, the threshold will be obtained for the minimal value of X (i. e. E^2) satisfying eq. (IV, 5).

$g(X, Y)$ is a continuous function of X and Y. Let us rewrite eq. (II, 30) and (IV, 4) as :

$$\begin{cases} \psi'(u) + 2\psi(u)(Y + X\cos^2 u) - \zeta^2 X\psi'_1 \cos u = 0 \\ \qquad\qquad\qquad\qquad\qquad\qquad\qquad (IV, 9) \\ \psi'_1 = \frac{1}{\pi} \int_0^{2\pi} \psi(u) \cos u \, du \qquad\qquad (IV, 10) \end{cases}$$

where $u = \omega t$ and $\psi'(u) = \partial\psi/\partial u$.

From these last equations it can be easily shown that
(i) When $X \to 0$, the condition for ψ to be periodic is

$$\lim_{X \to 0} \frac{1}{\zeta^2} = \lim_{X \to 0} g(X, Y) = \frac{YX}{4 Y^2 + 1} + 0(X^2) \ (Y \neq 0)$$

$$\lim_{X \to 0} \psi(u) = \left(\frac{2 Y\cos u + \sin u}{4 Y^2 + 1}\right)\psi'_1 .$$

Thus, for $X \to 0$, $g(X, Y) \to 0$.
(ii) When $X \to \infty$, from eq. (IV, 9), for finite Y, $\psi(u) \to \psi'_1\zeta^2/2 \cos u$ and from eq. (IV, 10)

$$\frac{1}{\zeta^2} = \lim_{X \to \infty} g(X, Y) = 1 .$$

(iii) If X or Y is negative, the integral in eq. (IV, 3) diverges.

Hence for any value of ζ^2 ($\zeta^2 > 1$) (⁵) one can find solutions of the equation

$$g(X, Y) = \frac{1}{\zeta^2} \quad (IV, 11)$$

and the curve defined by this equation is located in the first quadrant ($X, Y \geqslant 0$).

Let X_m and Y_m be the values of X and Y that give the absolute minimum of X on this curve. X_m and Y_m correspond to the threshold conditions and depend only on ζ^2. This leads to two requirements, listed below under (α) and (β) :

α) *Threshold field.*

$$\frac{E_M^2}{2} = \overline{E^2(t)} = \frac{X_m(\zeta^2)}{\lambda} \omega . \quad (IV, 12)$$

From eq. (IV, 12), $\overline{E^2(t)}$ appears to be proportional to ω. This is an asymptotic result for high ω. At slightly lower frequencies, the correct result is $\overline{E^2} = \text{constant} + \omega X_m/\lambda$. We shall now discuss briefly an improved approximation leading to this form.

We must take into account the fact that X_m depends not only on ζ^2 but also on $\omega\tau$. For $\omega\tau \gg 1$, we can expand X_m on powers of $1/\omega\tau$:

$$X_m(\zeta^2, \omega\tau) = X_m^0(\zeta^2) + \frac{1}{\omega\tau} X_m^1(\zeta^2) + 0(\omega^2 \tau^2) .$$

(⁵) It appears, from computer calculations, that, eq. (IV, 11) has no solution for $\zeta^2 < 1$. We have not been able to give a mathematical demonstration of this point. It implies that there is no « dielectric » instability for ($\zeta^2 - 1$) < 0.

Then we have

$$\overline{E^2(t)} = \frac{1}{\lambda}\left[\omega X_m^0(\zeta^2) + \frac{1}{\tau}X_m^1(\zeta^2)\right] \qquad \text{(IV, 13)}$$

and we find a straight line for the plot of $\overline{E^2}$ versus ω. This is in good agreement with the experimental result [5], [17], [18]. Finally, inserting eq. (II, 19) in eq. (IV, 13) one obtains

$$\overline{E^2(t)} = \left[\frac{\varepsilon_\parallel}{\varepsilon_\perp}\frac{4\pi}{\varepsilon_a}\frac{\gamma_1\eta_0}{(\gamma_1 + \eta_0)}X_m^0(\zeta^2)\right] \times$$

$$\times \left[\omega + \frac{X_m^1(\zeta^2)}{\tau X_m^0(\zeta^2)}\right]. \qquad \text{(IV, 14)}$$

It must be noted that, from eq. (IV, 14), the *field* threshold does not depend on the wave vector k : thus it is independent of sample thickness [5], [17], [18]. This is very different from the conduction regime ($\omega < \omega_c$) where the *voltage* threshold was independent of thickness. The existence of a field threshold for $\omega > \omega_c$ is well confirmed by the experiments [17].

β) Spatial periodicity of the molecular pattern at threshold. — At threshold we have

$$E_0^2 = \frac{2\,\omega}{\lambda}\,Y_m(\zeta^2, \omega\tau)\,.$$

This is an implicit equation for the wave vector k defining the spatial periodicity. Expanding, as previously, $Y_m(\zeta^2, \omega\tau)$ on powers of $1/\omega\tau$, one finds, for $\omega\tau \gg 1$,

$$E_0^2 = \frac{2}{\lambda}\left[\omega Y_m^0(\zeta^2) + \frac{1}{\tau}Y_m^1(\zeta^2)\right]. \qquad \text{(IV, 15)}$$

Inserting eq. (II, 4) and (II, 19), one obtains :

$$\chi_a\,H^2 + K_{33}\,k^2 =$$

$$= \frac{2\,\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\left[\omega Y_m^0(\zeta^2) + \frac{1}{\tau}Y_m^1(\zeta^2)\right]. \qquad \text{(IV, 16)}$$

Eq. (IV, 16) shows that, at threshold,

(i) for fixed H, k^2 is a linear function of ω
(ii) for fixed ω, the quantity $(\chi_a\,H^2 + K_{33}\,k^2)$ remains constant.

These two points have been confirmed by recent experiments on M B B A at Orsay [17].

In order to reach more quantitative comparisons between theory and experiment, we have numerically computed the function $g(X, Y)$ for various values of ζ^2 ($1 < \zeta^2 < 20$). On figures (3-4), $1/g(X, Y)$ is plotted versus $1/X$ in the range $X > 5$, $Y < 2.5$.

A more accurate calculation of Y_m^0 and X_m^0 has also been carried on a computer for values of ζ^2 corresponding to the experimental conditions.

One finds $X_m^0 = 1.04$, $Y_m^0 = 0.31$ for $\zeta^2 = 3.05$ and $X_m^0 = 0.58$, $Y_m^0 = 0.37$ for $\zeta^2 = 4.5$.

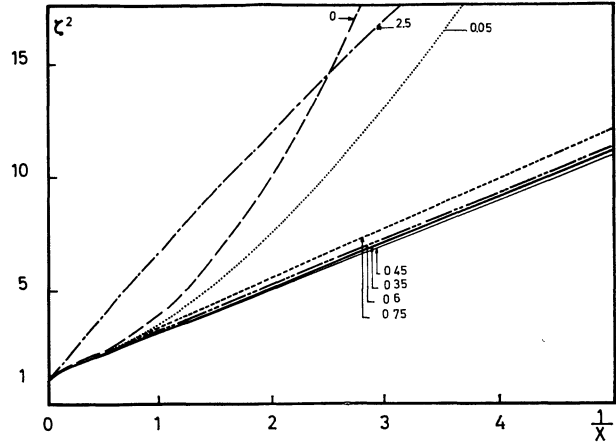A crude estimation of ζ^2 gives 3.2. The slope of the

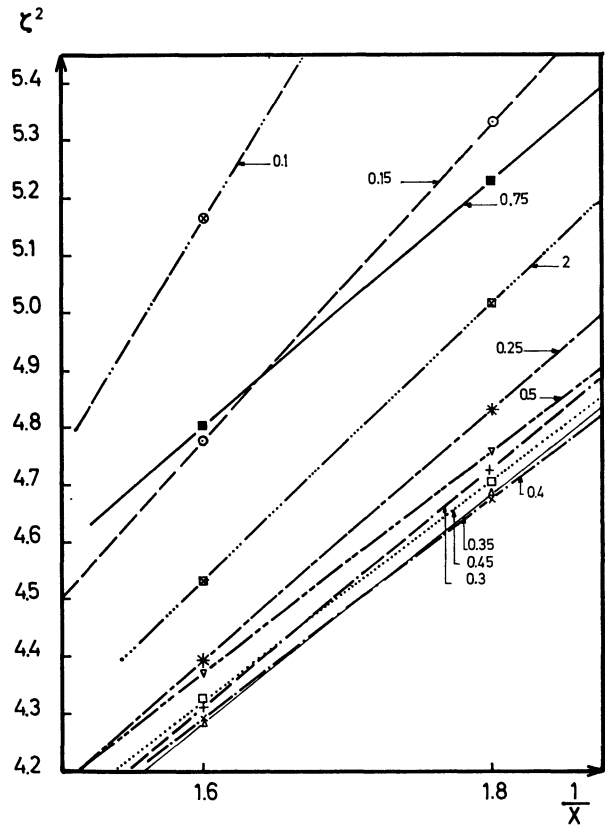FIG. 3. — ζ^2 is plotted versus $1/X$ for different values of the parameter Y.

FIG. 4. — Enlarged detail of figure 3. One can see how the curves are intercrossing for different values of Y.

experimental curve $\overline{E^2}(\omega)$ is in a good agreement with the one that can be deduced from the first set (X_m^0, Y_m^0) [17]. The agreement between theory and experiment is less good for the slope of $E_0^2(\omega)$. The slope of this curve depends on the conductivity of the material, and does not remain constant when ω increases. It appears [17] that the agreement between the theoretical and experimental slopes is reasonable only for small values of k^2. The discrepancy that occurs for high values of k^2 could be due to the effects of the diffusion currents (see Appendix A).

It must be emphasized that, in this regime, the space charge is time independent, but the curvature (and the molecular preferred orientation) has a rather complicated oscillation with period $2\,\pi/\omega$. $(\psi(t))^2$ is shown on figure 5. The local dielectric tensor oscillates with the same period. We can then legitimately call this regime a « dielectric regime ».
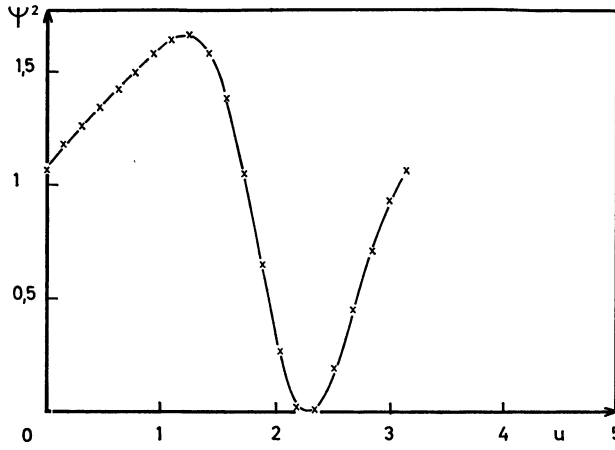

FIG. 5. — $\psi^2(u)$ is plotted versus u for : $u(0, \pi)$, $\zeta^2 = 3.05$ and
$$Y = Y_m^0 = 0.31\,.$$

V. The case $\varepsilon_\parallel > \varepsilon_\perp$. — By mixing suitable nematic components one can vary continuously the dielectric anisotropy ε_a and even change its sign : in the present section, we shall discuss the instability threshold as a function of ε_a, allowing ε_a to be either positive or negative. We make two restrictions however

a) even when $\varepsilon_a > 0$, we consider only the case where the unperturbed optical axis is *parallel to the walls* (along x, in the notation of this paper), the alignment being due either to a magnetic field H or to suitable boundary conditions at the walls

b) we restrict our attention to nematics with $\sigma_a > 0$ (no system with $\sigma_a < 0$ being known at present).

In spite of these two restrictions, we shall find a great variety of possible behaviors. The results are summarised in figure 6.

Our investigation will be mainly concerned with the « conduction regime », which will turn out to be often dominant here. The fundamental equation is still eq. (III, 9), which applies for both signs of ε_a. We may rewrite (III, 9) in a form where the role of the dielectric anisotropy is more apparent.

$$\overline{E^2} = D_0^2\,\frac{1 + \omega^2\tau^2}{\varepsilon_a\,\omega^2\,\tau^2 + \theta_H}\qquad (V, 1)$$

where D_0 is defined by eq. (II, 4), and the Helfrich parameter θ_H is given explicitly by :

$$\theta_H = -\,\varepsilon_a(\zeta^2 - 1) = \frac{\varepsilon_\parallel}{\sigma_\parallel}\left[\sigma_a\,\delta + \frac{\sigma_\perp}{\varepsilon_\perp}\,\varepsilon_a(1 - \delta)\right]\quad (V, 2)$$

δ is a dimensionless ratio involving the friction coefficients

$$\delta = \frac{2(\gamma_1 - \gamma_2)}{\gamma_1 + 2\,\alpha_4 + \beta - 2\,\gamma_2} =$$

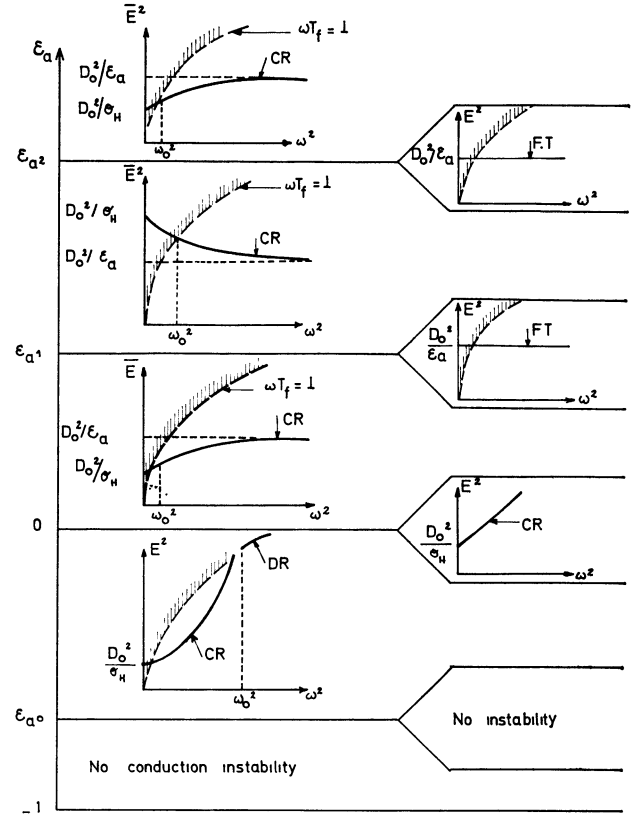$$= \frac{2\,\gamma_1}{\gamma_1 - \gamma_2}\,\frac{1}{1 + \eta_0/\gamma_1}\,.\qquad (V, 3)$$

FIG. 6. — Instability mean square threshold field versus ω^2 for different values of ε_a. The dielectric regime (D. R.) occurs only in the range $\varepsilon_{a_0} < \varepsilon < 0$. For $\varepsilon_a = \varepsilon_{a_1}$, ε_{a_2}, $\zeta^2 = 0$ and the Carr-Helfrich process disappear. D_0^2/ε_a is the threshold for the Fredericks transition.

From thermodynamic inequalities [16] γ_1, η_0, and $\gamma_1 - \gamma_2$ are positive. From the experimental data on alignment by flow in nematics Leslie [13] has concluded that $-\,\gamma_2 \geqslant \gamma_1$. These remarks lead to the inequalities $0 < \delta < 1$. For P A A we expect $\delta \simeq 0.7$ (and this order of magnitude also applies probably to M B B A). Note that ζ^2 (in eq. V, 2) may now be positive or negative. Some special values of ζ^2 play a crucial role in the discussion

a) $\zeta^2 = 1$. This is obtained when $\varepsilon_a = \varepsilon_{a0}$, where

$$\varepsilon_{a0} = -\,\frac{\delta}{1 - \delta}\,\frac{\sigma_a}{\sigma_\perp}\,\varepsilon_\perp < 0\,.\qquad (V, 4)$$

For $\varepsilon_a < \varepsilon_{a0}$ the nematic is absolutely stable under fields of arbitrary strength and frequency [6]

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[6] The value ε_{a0} defined by (V, 4) is acceptable only if it corresponds to values of $\varepsilon_\parallel$ and $\varepsilon_\perp$ which are both positive : this requires $\sigma_a/\sigma_\perp < (1 - \delta)/\delta$.

b) $\zeta^2 = 0$. This may occur for two distinct values of ε_a [7]

$$\left.\begin{array}{l} \varepsilon_{a1} = \varepsilon_{\parallel} \, \sigma_a/\sigma_{\parallel} \\ \varepsilon_{a2} = \varepsilon_{\parallel} \, \delta \end{array}\right\} \; (> 0) \, . \qquad (V, 5)$$

For $\zeta^2 = 0$ the torques due to the Carr Helfrich effect vanish exactly [8] ; eq. (V, 1) gives a threshold independent of frequency $E^2 = D_0^2/\varepsilon_a$. This could have been derived from the standard discussion of the competition between the field H — plus the effect of the walls — (favoring an alignment along x) and a static field $\sqrt{\overline{E^2}}$ (favoring an alignment along z).

In the cases which have been studied experimentally we have $\varepsilon_{a1} < \varepsilon_{a2}$. We shall restrict our attention to this case in what follows.

Finally, returning to eq. (V, 1), we find the following types of behavior upon increasing ε_a :

1) $\varepsilon_a \leqslant \varepsilon_{a0}$ = complete stability

2) $\underline{\varepsilon_{a0} < \varepsilon_a < 0}$ $\begin{cases} \text{conduction regime } \omega < \omega_c \\ \text{dielectric regime } \omega > \omega_c \end{cases}$

(this is the case discussed in the previous sections).

3) $\underline{\varepsilon_a \geqslant 0}$ conduction regime at all frequencies [9]. Here we have three sub-classes

a) $0 \leqslant \varepsilon_a < \varepsilon_{a1}$ $\overline{E}$ increasing function of ω
b) $\varepsilon_{a1} < \varepsilon_a < \varepsilon_{a2}$ $\overline{E}$ decreasing function of ω
c) $\varepsilon_{a2} < \varepsilon_a$ $\overline{E}$ increasing function of ω

These results are summarised on fig. (6). On this figure we have also delineated the region in which the approximation $\omega T_f \gg 1$ is valid, since this approximation underlies eq. (V, 1). It is clear from the figure that, in case 3), the conduction regime may indeed extend to arbitrarily high frequencies [9]. Some preliminary results on this case have been obtained recently at Freiburg [19].

VI. Limitations of this model.

In this model, we have considered the Helfrich-Carr instabilities for an AC electric field in an infinite medium ; the boundary conditions at the surface of the sample have been included in a very approximate manner. This limits the validity of the model and may explain some discrepancies with the experimental results. Some other discrepancies may be due to the existence of diffusion currents that we have neglected. Nethertheless it appears that this model is able to explain most of the experimental results for materials having a negative dielectric anisotropy and to give some predictions for the case of a positive dielectric anisotropy.

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[7] To derive eq. (V, 5) it is convenient to write $\zeta^2 = \sigma_H \, \tau/\lambda\eta$ (eq. III, 8). The two roots correspond to the zeros of σ_H and of $1/\eta$.

[8] This is shown in appendix C.

[9] Of course, at some frequency ω_1 the dispersion of the dielectric constant would become important : typically we will be in the range of 10^5 cycles.

Provided $d/d_c \gg 1$, this model is valid for the whole frequency range of interest $(0 - 10^5 \, \text{Hz})$ except in two small frequency intervals.

a) for very low frequencies $(\omega \lesssim \omega_0)$ where the assumption $\omega T_f \gg 1$ does not hold. However, for very low frequencies, the instabilities may be due to carrier injection (such as in « Felici instabilities) » and not to the Carr-Helfrich process ;

b) for frequencies of the order of the « cut-off » frequency ω_c. In this range the instability regime is neither a conduction regime nor a dielectric regime, but can be considered as a mixing of both, in agreement with observations made at Orsay.

APPENDIX A : Contribution of the diffusion currents.

We have neglected in section (II, 2) the diffusion currents. We will now take into account these currents and show that their contribution can be neglected.

Let us call q_i the contribution of ion i to the excess-charge and $\overline{\overline{D}}_i$ its diffusion tensor

$$D_i = D_{i\perp} + D_{ia} \, (\mathbf{nn}) \qquad (A, 1)$$

The total electric current is

$$\mathbf{J}' = \mathbf{J} - \sum_i \overline{\overline{D}}_i \cdot \mathbf{grad} \, q_i \qquad (A, 2)$$

when $\mathbf{J}$ is defined by eq. (II,7). The charge-balance equation is now

$$\dot{q} + \frac{q}{\tau} - \sum_i D_{i\parallel} \frac{\partial^2 q_i}{\partial x^2} + \sigma_H E\psi = 0 \, . \qquad (A, 3)$$

Let us define the average diffusion constant as

$$D_{\parallel} = \left(\sum_i D_{i\parallel} \, q_i\right) \Big/ \left(\sum_i q_i\right)$$

and the Debye screening parameter

$$q_D^2 = \frac{1}{\tau D_{\parallel}} \, .$$

Eq. (A, 3) can be rewritten as

$$\dot{q} + \frac{q}{\tau'} + \sigma_H E\psi = 0 \qquad (A, 4)$$

with

$$\frac{1}{\tau'} = \frac{1}{\tau}\left(1 + \frac{k^2}{q_D^2}\right) \, . \qquad (A, 5)$$

For M B B A, $D \simeq 10^{-7}$ cgs ; $1/\tau \simeq 4 \times 10^2 \, \text{s}^{-1}$;

$q_D^2 \simeq 4 \times 10^9\ \mathrm{cm}^{-2}$. In the conduction regime, $k \simeq \pi/d$, and taking $d = 100\ \mu$, $k^2 \simeq 10^3\ \mathrm{cm}^{-2}.k^2/q_D^2$ can be neglected.

In the dielectric regime for frequencies of the order of 400 Hz, one finds q^2 in the range $2 - 4 \times 10^8\ \mathrm{cm}^{-2}.k^2/q_D^2$ can still be neglected.

However it must be emphasized that, for higher frequencies, leading to higher values of k^2, one could not neglect these terms. In section IV, $1/\zeta^2$ must be replaced by $1/\zeta^2\ (1 + k^2/q_D^2)$. Therefore the results of section IV are no longer valid : $E_2^0\ (\omega)$ and $E^2(\omega)$ are not straight lines, and, for ω fixed, $(\chi_a\,H^2 + K_{33}\,k^2)$ is not constant in the range $k^2 \simeq q_D^2$.

APPENDIX B : Extension of the Bloch-Floquet theorem. — The proof of eq. (II, 32) can be set on the following lines :

(i) The system of equations

$$\begin{aligned} \dot{y} + f(t)\,y + g(t)\,z &= 0 \\ \dot{z} + h(t)\,y + k(t)\,z &= 0 \end{aligned} \qquad \text{(B, 1)}$$

can be written as

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\widehat{\psi}(t) = - \big(M(t)\big)\,\widehat{\psi}(t) \qquad \text{(B, 2)}$$

where $\widehat{\psi}(t)$ is a spinor with components $(y(t), z(t))$ and $\big(M(t)\big)$ a square matrix

$$\big(M(t)\big) = \begin{bmatrix} f(t) & g(t) \\ h(t) & k(t) \end{bmatrix}.$$

The general solution is

$$\widehat{\psi}(t) = a_1\,\widehat{\psi}_1(t) + a_2\,\widehat{\psi}_2(t) \qquad \text{(B, 3)}$$

where $\widehat{\psi}_1(t)$ and $\widehat{\psi}_2(t)$ are linearly independent solutions (and a_1 and a_2 are time-independent) :

$$\widehat{\psi}_1(t) = \mathcal{A}(t)\,\widehat{\psi}_1(0)\,.$$

Using the Feynman conventions for the ordering of non commuting operators, one can write the evolution operator $\mathcal{A}(t)$ as

$$\mathcal{A}(t) = \exp - \int_0^t \big(M(t)\big)\,\mathrm{d}t\,.$$

(ii) Let $\big(M(t)\big)$ have period T. If $\widehat{\psi}(t)$ is a solution of eq. (B, 2), $\widehat{\psi}(t + T)$ is also a solution of this equation. We can thus write

$$\begin{aligned} \widehat{\psi}_1(t + T) &= a_{11}\,\widehat{\psi}_1(t) + a_{12}\,\widehat{\psi}_2(t) \\ \widehat{\psi}_2(t + T) &= a_{21}\,\widehat{\psi}_1(t) + a_{22}\,\widehat{\psi}_2(t) \end{aligned} \qquad \text{(B, 4)}$$

where the a's are time-independent constants.

(iii) We can find two linearly independent spinors $\widehat{\psi}_i\ (i = 1,2)$ such as

$$\widehat{\psi}_i(t + T) = \lambda_i\,\widehat{\psi}_i(t) \quad (i = 1,2) \qquad \text{(B, 5)}$$

where λ_1 and λ_2 are the roots of the secular equation

$$\mathrm{Det}\,|\,a_{ji} - \lambda\delta_{ij}\,| = 0 \qquad \text{(B, 6)}$$

State

$$\lambda_i = \mathrm{e}^{s_i T}\,;\,\widehat{\varphi}_i(t) = \mathrm{e}^{-s_i t}\,\widehat{\psi}_i(t)\,.$$

From eq. (B, 5), the spinor $\widehat{\varphi}_i(t)$ is periodic with period T. This proves eq. (II, 32).

APPENDIX C : Torque balance in the conduction regime. — Let us rewrite eq. (II, 24) as

$$\frac{\partial \Gamma_{\mathrm{tot}}}{\partial x} = \frac{\partial}{\partial x}\,(\Gamma_F + \Gamma_H) \qquad \text{(C, 1)}$$

where

$$\frac{\partial \Gamma_F}{\partial x} = \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\,\lambda\left(E^2 - \frac{D_0^2}{\varepsilon_a}\right) \qquad \text{(C, 2)}$$

or

$$\frac{\partial \Gamma_F}{\partial x} = \left(\chi_a\,H^2 + K_{33}\,k^2 - \frac{\varepsilon_a}{4\,\pi}\,\frac{\varepsilon_\perp}{\varepsilon_\parallel}\,E^2\right)\psi \qquad \text{(C, 3)}$$

and

$$\frac{\partial \Gamma_H}{\partial x} = \frac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\left(\dot{\psi} + \frac{qE}{\eta}\right). \qquad \text{(C, 4)}$$

Γ_F is the part of the torque (independent of the space-charge) which is taken into account in the standard discussion of Fredericks transition. Γ_H is the contribution of the Carr-Helfrich process to the torque.

In the « conduction regime », the relaxation time T_f for the molecular orientation is much longer than the period of the electric field. Hence we may consider only the average values (over one period) of the torque. Then we have

$$\begin{cases} \Gamma_F = \dfrac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\,\lambda(\overline{E^2} - D_0^2/\varepsilon_a)\,\varphi & \text{(C,5)} \\[3mm] \Gamma_H = -\left(\dfrac{\gamma_1\,\eta_0}{\gamma_1 + \eta_0}\,\lambda\right)\dfrac{\zeta^2}{1 + \omega^2\,\tau^2}\,\overline{E^2}\,\varphi\,. & \text{(C, 6)} \end{cases}$$

From eq. (C, 6), Γ_H vanishes if ζ^2 vanishes.

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