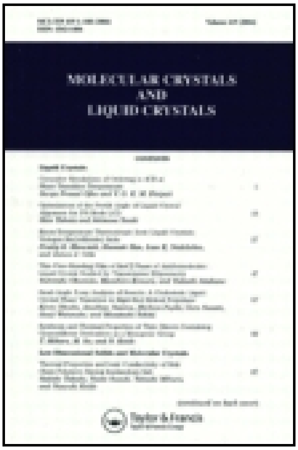

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Electrohydrodynamic Instability Limits of Nematics

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Electrohydrodynamic Instability Limits of Nematics

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A linear, two-dimensional formulation of the problem of the electrohydrodynamic instability limits of nematic liquid crystals is presented. The nematic is assumed to have negative dielectric anisotropy, homogeneous alignment, and to be excited by a square-pulsed electric field. The formulation enables one to determine, using digital computational techniques, the limiting values of the electric field for the instability and the respective spatial wavenumbers of the nematic director, at both, the conductivity and the dielectric, regimes. The method consists, essentially, in solving a system of two equations in terms of the applied electric field and the director wavenumber, so to retain, among the solutions, those giving the maximum and minimum values of the field.

The program was applied to the case of the room temperature nematic *N*-(*p*-Methoxybenzylidene)-*p*-Butylaniline (MBBA). The results are found to be in good agreement with the known experimental facts.

I INTRODUCTION

The electrohydrodynamic (EHD) instabilities exhibited by negative dielectric anisotropy nematic liquid crystals (NLC), under the action of d.c. or periodic electric fields,[1] have been the subject of a number of theoretical works.[2–9] Many of them, and in particular those of Refs. 2, 4 and 6, contributed much to the understanding of the instability mechanism. Nevertheless, and owing to the mathematical complexity of the problem, all of these theories were, in one or another way, approximative,[4–7] and/or restricted to the d.c. case. We mention, by the way, a recent numerical analysis work,[10] based on the one-dimensional theory of Refs. 4 and 6.

This paper is to present a linear two-dimensional formulation of the problem of the EHD instability limits in a NLC, by which one is capable, using digital computational techniques, to determine the limiting values of the electric field for the instability, and the respective director spatial wavenumbers (DSW), assuming the NLC under the action of a square-pulsed electric field.

II SETTING UP THE EQUATIONS

We consider the NLC as a conducting dielectric material placed between the parallel plates of a capacitor, the plate dimensions of which are much greater than the plate separation L (usually 10–500 microns). The initial direction of director's orientation is assumed to be parallel to the capacitor plates and the same everywhere, defining the x-axis. (Experimentally, this is achieved by a constant magnetic field or by properly rubbing the inner surfaces of the capacitor plates). The direction normal to the plates is defined as the z-axis. A voltage $V(t)$ is supposed to be applied to the capacitor (Figure 1). We want

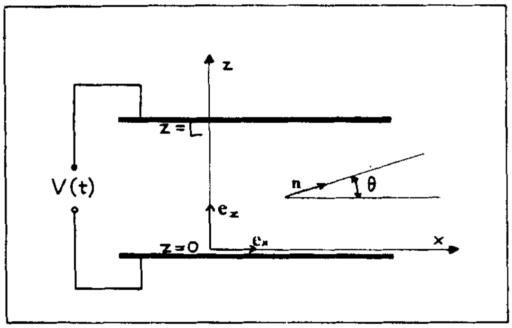

FIGURE 1 The geometry of the NLC cell.

to determine the limiting effective values of $V(t)$ necessary to excite the NLC to EHD instability. Following all the previously mentioned works, we assume that no charge injection process is taking place at the electrodes. This has been proved to be achievable experimentally.[1] In this way we restrict ourselves to the nematics' EHD response only.[1]

In setting up the equations, all variables are assumed sufficiently small, so that, we neglect all terms of higher than the first order, having as a result, linear equations only. This is not, in fact, an approximation because our task is the determination of the limiting (instability) condition only.

From the geometry, as it has been defined, it is apparent that the motion of the director, and also of the nematic liquid as a bulk, close to the limiting condition, will be planar, parallel to the x-z plane. Thus the director will be given by:

$$\mathbf{n} = \mathbf{e}_x + \theta \mathbf{e}_z, \quad |\theta| \ll 1 \tag{1}$$

θ being the angle between the director and the x-axis in the disturbed state (Figure 1).

Adopting Leslie's continuum-mechanical theory on the anisotropic liquids (assumed incompressible),[11,12] and neglecting inertial terms,[4,13] we take the following equations:

a) Conservation of linear momentum along x-axis:

$$-\frac{\partial p}{\partial x} + (\eta_3 - \eta_1 - \alpha_2)\frac{\partial^2 \upsilon_x}{\partial x^2} + (\eta_1 + \gamma_2)\frac{\partial^2 \upsilon_x}{\partial z^2} + \alpha_3 \frac{\partial \dot{\theta}}{\partial z} = 0 \tag{2}$$

where

p:	The pressure inside the NLC
υ_x:	The x-component of the flow velocity
$\alpha_1, \ldots, \alpha_6$:	Leslie's viscosity coefficients[11] connected by the relation:[14]

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

$$\eta_1 \equiv (\alpha_4 + \alpha_5 - \alpha_2)/2$$

$$\eta_3 \equiv \alpha_1 + \alpha_4 + \alpha_5 + \alpha_6, \quad \gamma_2 \equiv \alpha_2 + \alpha_3$$

b) Conservation of linear momentum along z-axis:

$$qE_z - \frac{\partial p}{\partial z} + \eta_1 \frac{\partial^2 \upsilon_z}{\partial x^2} + (\alpha_4 - \eta_1 - \alpha_2)\frac{\partial^2 \upsilon_z}{\partial z^2} + \alpha_2 \frac{\partial \dot{\theta}}{\partial x} = 0 \tag{3}$$

where

E_z:	The z-component of the electric field
q:	The excess charge density
υ_z:	The z-component of the flow velocity

Using, furthermore, the continuity relation for the incompressible fluids:

$$\frac{\partial \upsilon_x}{\partial x} + \frac{\partial \upsilon_z}{\partial z} = 0 \tag{4}$$

and eliminating p and υ_x between (2), (3) and (4), we get

$$E_z \frac{\partial^2 q}{\partial x^2} + \left(\alpha_2 \frac{\partial^2}{\partial x^2} - \alpha_3 \frac{\partial^2}{\partial z^2}\right)\dot{\psi} + \left\{\eta_1 \frac{\partial^4}{\partial x^4} + \eta_2 \frac{\partial^2}{\partial x^2}\frac{\partial^2}{\partial z^2} + (\eta_1 + \gamma_2)\frac{\partial^4}{\partial z^4}\right\}\upsilon_z = 0 \tag{5}$$

where

$$\eta_2 \equiv \alpha_1 + \alpha_3 + \alpha_4 + \alpha_5, \quad \psi \equiv \partial\theta/\partial x$$

c) Adopting Frank's expression for the curvature elasticity free energy density of the NLC,[15,16] we get the following equation for the angular momentum conservation along y-axis:

$$\gamma_1 \dot{\psi} - \left(K_{11}\frac{\partial^2}{\partial z^2} + K_{33}\frac{\partial^2}{\partial x^2}\right)\psi - \frac{\varepsilon_\alpha E_z}{4\pi}\frac{\partial E_x}{\partial x} + \left(\chi_\alpha H^2 - \frac{\varepsilon_\alpha E_z^2}{4\pi}\right)\psi + \left(\alpha_2\frac{\partial^2}{\partial x^2} - \alpha_3\frac{\partial^2}{\partial z^2}\right)v_z = 0 \quad (6)$$

where

$\gamma_1 \equiv \alpha_3 - \alpha_2$	
K_{11}, K_{33}:	Frank's splay and bend elastic constants, respectively.
χ_α:	The anisotropy of the magnetic susceptibility, assumed positive.
$\varepsilon_\alpha = \varepsilon_p - \varepsilon_n$:	The dielectric anisotropy
$\varepsilon_p, \varepsilon_n$:	The dielectric constant along and normal to the director, respectively.
H:	The magnetic field, assumed parallel to the x-axis.
E_x:	The x-component of the electric field.

In deriving the Eq. (6) use has been made of the continuity relation (4).

d) The z-component of the electric field may be considered as a composition of two terms:[4] A coordinate independent term

$$E = V(t)/L$$

and a coordinate dependent one

$$E_z' \equiv E_z - E, \quad |E_z'| \ll |E|$$

From the electrostatic relation $\nabla \times \mathbf{E} = 0$ we get

$$\frac{\partial E_z'}{\partial x} = \frac{\partial E_x}{\partial z} \quad (7)$$

The dielectric tensor is given by

$$\bar{\bar{\varepsilon}} = \varepsilon_\alpha \mathbf{nn} + \varepsilon_n \bar{\bar{I}} \quad (8)$$

$\bar{\bar{I}}$ being the unit tensor. The Gauss' law

$$\nabla \cdot (\bar{\bar{\varepsilon}} \cdot \mathbf{E}) = 4\pi q$$

combined with the equations (1), (7) and (8), gives

$$\left(\varepsilon_p \frac{\partial^2}{\partial x^2} + \varepsilon_n \frac{\partial^2}{\partial z^2}\right)E_x + \varepsilon_\alpha E \frac{\partial \psi}{\partial x} = 4\pi \frac{\partial q}{\partial x} \tag{9}$$

The conductivity and diffusion tensors are given, respectively, by

$$\bar{\bar{\sigma}} = \sigma_\alpha \mathbf{nn} + \sigma_n \bar{\bar{I}} \tag{10}$$

$$\bar{\bar{D}} = D_\alpha \mathbf{nn} + D_n \bar{\bar{I}} \tag{11}$$

where

$\sigma_\alpha \equiv \sigma_p - \sigma_n$:	The anistropy of the conductivity (assumed to be positive), where
σ_p, σ_n:	The conductivity of the NLC along and normal to the director, respectively.
$D_\alpha \equiv D_p - D_n$:	The anisotropy of the diffusion constant.
D_p, D_n:	The diffusion constants along and normal to the director, respectively.

(Here is implicit the approximation of equating the diffusion constants of all kinds of ions present)

The current density is given by

$$\mathbf{j} = \bar{\bar{\sigma}} \cdot \mathbf{E} - \bar{\bar{D}} \cdot \nabla q \tag{12}$$

Applying charge conservation, $\nabla \cdot \mathbf{j} + \dot{q} = 0$, and using (1), (7), (10), (11) and (12) we get

$$\left(\sigma_p \frac{\partial^2}{\partial x^2} + \sigma_n \frac{\partial^2}{\partial z^2}\right)E_x - \left(D_p \frac{\partial^2}{\partial x^2} + D_n \frac{\partial^2}{\partial z^2}\right)\frac{\partial q}{\partial x} + \frac{\partial \dot{q}}{\partial x} + \sigma_\alpha E \frac{\partial \psi}{\partial x} = 0 \tag{13}$$

With the Carr–Helfrich model of EHD instability[2] in mind, we postulate that the functions q, ψ and $\partial E_x/\partial x$ have the same spatial dependence which, according to (9), (and assuming also separability of the variables x and z), must have the form

$$\cos(kx + \phi_x)\sin(mz + \phi_z) \tag{14}$$

The boundary condition

$$E_x = 0 \quad \text{at } z = 0 \text{ and at } z = L$$

gives

$$\phi_z = 0, \quad m = n\pi/L, \quad n = 1, 2, \ldots. \tag{15}$$

The k in (14) is, of course, the DSW.

Eliminating, now, E_x between (9) and (13), and using (14) we get

$$\dot{q} + a_1 q + b_1 E\psi = 0 \tag{16}$$

e) Finally eliminating E_x and v_z between (5), (6) and (9), and using again (14) we find

$$\dot{\psi} + a_2\psi + b_2 Eq = 0 \tag{17}$$

By defining

$$\begin{aligned}
\xi &\equiv k^2 + m^2 \\
\xi_\varepsilon &\equiv \varepsilon_p k^2 + \varepsilon_n m^2 \\
\xi_\sigma &\equiv \sigma_p k^2 + \sigma_n m^2 \\
\xi_D &\equiv D_p k^2 + D_n m^2 \\
\xi_K &\equiv K_{33} k^2 + K_{11} m^2 \\
\xi_\alpha &\equiv -\alpha_2 k^2 + \alpha_3 m^2 \\
\xi_\eta &\equiv \eta_1 k^4 + \eta_2 k^2 m^2 + (\eta_1 + \gamma_2) m^4
\end{aligned}$$

the parameters a_1, a_2, b_1 and b_2 of equations (16) and (17) are given by the relations:

$$\begin{aligned}
a_1 &= 4\pi\xi_\sigma/\xi_\varepsilon + \xi_D \\
b_1 &= (\varepsilon_n\sigma_p - \varepsilon_p\sigma_n)\xi/\xi_\varepsilon \\
a_2 &= \{\chi_a H^2 + \xi_K - \varepsilon_a\varepsilon_n E^2\xi/(4\pi\xi_\varepsilon)\}/(\gamma_1 - \xi_\alpha^2/\xi_\eta) \\
b_2 &= k^2(\xi_\alpha/\xi_\eta - \varepsilon_a/\xi_\varepsilon)/(\gamma_1 - \xi_\alpha^2/\xi_\eta)
\end{aligned}$$

The system of equations (16) and (17), in the case where $m = 0$, reduces (aside from notation) to the respective system of the one-dimensional theory.[4,6]

III SQUARE-PULSE FIELDS

For a field of the form:

$$e(t) = E, \quad t > 0$$

the system (16)–(17) has the solution:

$$\begin{pmatrix} q(t) \\ \psi(t) \end{pmatrix} = \begin{pmatrix} \lambda_2(t) & -\mu_1(t) \\ -\mu_2(t) & \lambda_1(t) \end{pmatrix} \begin{pmatrix} q(0) \\ \psi(0) \end{pmatrix} \tag{18}$$

where

$$\begin{aligned}
\lambda_i(t) &\equiv \{(s_1 + a_i)\exp(s_1 t) - (s_2 + a_i)\exp(s_2 t)\}/D \quad i = 1, 2 \\
\mu_i(t) &\equiv b_i E\{\exp(s_1 t) - \exp(s_2 t)\}/D, \quad i = 1, 2 \\
s_1 &\equiv -(a_1 + a_2)/2 + D/2 \\
s_2 &\equiv -(a_1 + a_2)/2 - D/2 \\
D &\equiv \{(a_1 - a_2)^2 + 4b_1 b_2 E^2\}^{1/2}
\end{aligned}$$

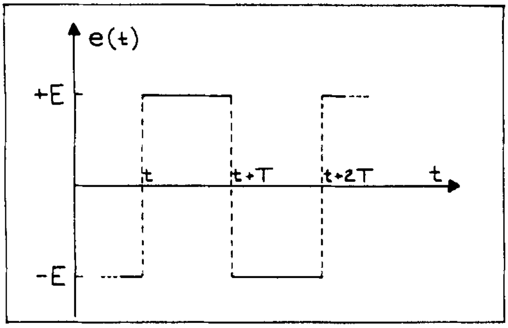

FIGURE 2 Time plot of the exciting the NLC electric field.

Applying, now, the solution (18) to the square-pulse field of Figure 2, and setting

$$\lambda_i \equiv \lambda_i(T), \quad i = 1, 2$$
$$\mu_i \equiv \mu_i(T), \quad i = 1, 2$$

where T is the half-period of the field, we find:

$$\begin{pmatrix} q(t + 2T) \\ \psi(t + 2T) \end{pmatrix} = \{A\} \begin{pmatrix} q(t) \\ \psi(t) \end{pmatrix} \tag{19}$$

where

$$\{A\} = \begin{pmatrix} \lambda_2^2 - \mu_1\mu_2 & \mu_1(\lambda_1 - \lambda_2) \\ \mu_2(\lambda_2 - \lambda_1) & \lambda_1^2 - \mu_1\mu_2 \end{pmatrix} \tag{20}$$

In the steady state, and for some integer β, we must have

$$\begin{pmatrix} q(t + \beta 2T) \\ \psi(t + \beta 2T) \end{pmatrix} = \begin{pmatrix} q(t) \\ \psi(t) \end{pmatrix} \tag{21}$$

From Eqs. (19) and (21) we infer that the β-th power of the matrix $\{A\}$ must have an eigenvalue equal to unity, and thus the matrix $\{A\}$ itself an eigenvalue of modulus unity. Now, the characteristic equation of $\{A\}$ is:

$$r^2 - G_1 r + G_2 = 0 \tag{22}$$

where

$$G_1 \equiv 2p_1p_2 + R^2(p_1 - p_2)^2$$
$$G_2 \equiv p_1^2 p_2^2$$
$$p_i \equiv \exp(s_i T), \quad i = 1, 2$$
$$R^2 \equiv (a_1 - a_2)^2/D^2$$

One may easily see that for a negative dielectric anisotropy NLC, and for all the cases of experimental interest ($k \gtrsim m$), the quantity D^2 is positive, and thus R^2 also positive. The discriminant of the Eq. (22) is equal to:

$$R^2(p_1 - p_2)^2\{R^2(p_1 - p_2)^2 + 4p_1p_2\}$$

and, thus, is positive. Therefore, Eq. (22) has two unequal, real, positive roots and, consequently, one of them must be equal to $+1$. So, from (22), we have:

$$G_2 + 1 = G_1$$

or, recalling the definitions following (22), and also the definitions of s_1 and s_2, we get

$$D \sinh\left(\frac{a_1 + a_2}{4f}\right) = u(a_2 - a_1)\sinh\left(\frac{D}{4f}\right), \quad u = \pm 1 \tag{23}$$

where $f \equiv 1/2T$ is the frequency of the applied field. Evidently

$$u = -1 \quad \text{implies} \quad a_1 > a_2 \tag{24}$$

$$u = +1 \quad \text{implies} \quad a_1 < a_2 \tag{25}$$

For every frequency value f, the extreme values of E for which either of (23) (considered as equations with unknown the k), have a solution, are the limiting field values for the EHD instability. The solution of (23) for these limiting values is the DSW on the limiting condition.

IV RESULTS

Setting

$$Q_u \equiv D \sinh\left(\frac{a_1 + a_2}{4f}\right) - u(a_2 - a_1)\sinh\left(\frac{D}{4f}\right)$$

the instability condition becomes

$$Q_u = 0, \quad u = \pm 1 \tag{26}$$

Either of (26) may be considered as an implicit function

$$E = E(k) \tag{27}$$

Differentiating (26) along the curve defined by (27)

$$dQ_u = \left(\frac{\partial Q_u}{\partial k} + \frac{\partial Q_u}{\partial E}\frac{dE}{dk}\right)dk = 0, \quad u = \pm 1 \tag{28}$$

and remembering that we are looking for the extrema of (27):

$$\frac{\mathrm{d}E}{\mathrm{d}k} = 0, \quad \frac{\mathrm{d}^2E}{\mathrm{d}k^2} \neq 0$$

we take from (28) the condition:

$$\frac{\partial Q_u}{\partial k} = 0, \quad u = \pm 1 \tag{29}$$

Solving, therefore, the system of equations (26) and (29) one finds a set of (E, k) pairs from which has to select (for each of the cases $u = \pm 1$) those corresponding to the absolute maximum and absolute minimum of (27).

Following the outlined procedure, the problem is solved in terms of the variables

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

in place of E and k respectively. Taking advantage of the form of (23) and the definitions of a_1, a_2, b_1, b_2 and D, we replaced the quantities $\sigma_p, \sigma_n, D_p, D_n, f$ and H by

$$\sigma_r \equiv \sigma_p/(K_{33}m^2)$$
$$\sigma_r' \equiv \sigma_n/(K_{33}m^2)$$
$$D_r \equiv D_p/K_{33}$$
$$D_r' \equiv D_n/K_{33}$$
$$f_r \equiv f/(K_{33}m^2)$$
$$H_r \equiv H/(K_{33}m^2)^{1/2}$$

respectively, reducing, thus, the number of needed parameters by three, because now the results are independent of K_{33}, L and the integer n of (15), about which we will have to say more in the future.

To obtain definite results use has been made of the following values for the constants of the NLC which correspond to the room temperature nematic MBBA.

$\alpha_1 = 0.07$ P, $\alpha_2 = -0.78$ P, $\alpha_3 = -0.01$ P, $\alpha_4 = 0.83$ P, $\alpha_5 = 0.46$ P, Ref. 17
$K_{11}/K_{33} = 0.8$, $K_{33} = 0.7 \times 10^{-6}$ dynes, Ref. 18
$\varepsilon_p = 4.72$, $\varepsilon_n = 5.25$, Ref. 19
$D_p = 0.5 \times 10^{-7}$ sec/cm^2, Ref. 20
$D_n = 0.33 \times 10^{-7}$ sec/cm^2, (our estimation)
$\sigma_p/\sigma_n = 1.5$, Ref. 19

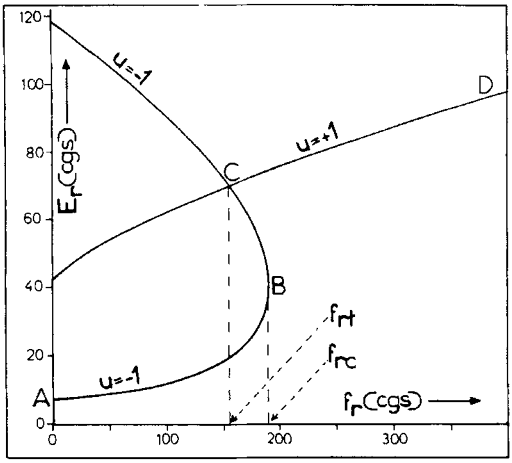

FIGURE 3 Calculated plot of the limiting "field" E_r versus "frequency" f_r for both, the conductivity and the dielectric, regimes, using $\sigma_r = 500$ cgs, $H_r = 0$. The region restricted by the curves ABC and CD and the f_r axis, is the stability region of the nematic.

Figures 3, 4 and 5 are to illustrate the results of the computer calculations, where use has been made of the values

$$\sigma_r = 500 \text{ cgs}, \quad H_r = 0$$

Figure 3 is a plot of the limiting "field" E_r versus "frequency" f_r for the cases $u = \pm 1$. One sees that the case $u = +1$ correspond to the dielectric

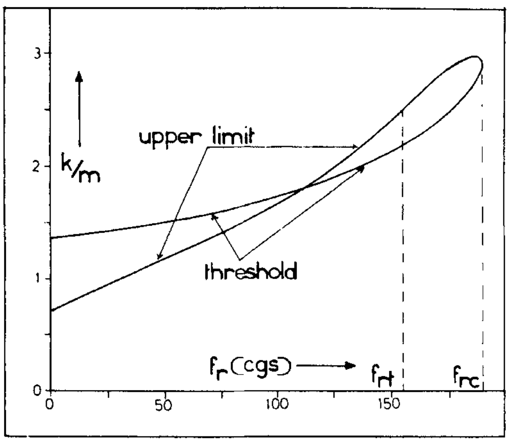

FIGURE 4 Calculated plot of threshold and upper limit "wavenumbers" k/m versus "frequency" f_r in the conductivity regime.

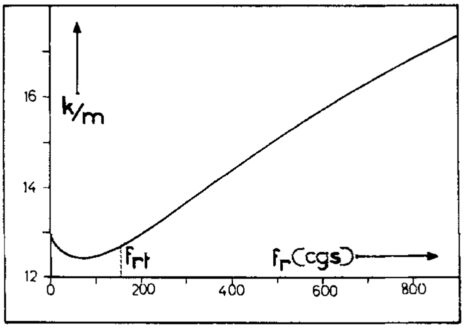

FIGURE 5 Calculated plot of the threshold "wavenumbers" k/m versus "frequency" f_r in the dielectric regime.

regime, whereas $u = -1$ to the conductivity regime.[1,4,6,21] One may connect this nomenclature with the inequalities (25) and (24) respectively. It is observed from the graph that, both regimes extend down to zero frequency in satisfactory accord to Ref. 22. The region limited by the curves ABC and CD and the "frequency" axis is the stability region of the NLC.[1,21] The quantities f_{rc} and f_{rt} correspond to the cut-off and transition frequencies, respectively.[6]

Figure 4 is a plot of the "frequency" dependence of the DSW divided by m, both, on the threshold and on the upper limit, of the conductivity regime. The threshold part of the curve may be compared to the respective experimental curves of Refs. 23 (Figure 5) and 24 (Figure 1), which, however, have been obtained under sinusoidal excitation. For the upper limit part of the curve no experimental work, for comparison reasons, have been found yet, and we are working on this line.

Finally, Figure 5 is a plot of the "frequency" dependence of the DSW, divided by m, on the dielectric threshold. The curve is not characterized by any frequency limit. Nevertheless, for sufficiently high frequencies, the relation (23), from which the curve of Figure 5 is obtained, have to be modified, because of the importance of the inertial and dielectric relaxation phenomena. However, up to 2000 Hz, the curve does not contradict our experimental results.

The upper limit DSW of the conductivity regime (Figure 4), and the dielectric DSW (Figure 5), for frequencies smaller than that of the transition value, is difficult to measure experimentally, because of the persisting dynamic scattering effects.[25] Probably, such measurements may be made with the NLC reported in Ref. 22.

A full exposition of the results of the presented theory concerning the dependence of the instability curves on the magnetic field, the conductivity and the thickness of the NLC cell, will be reported in a future paper.

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