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ELECTROHYDRODYNAMIC INSTABILITIES IN THE LIQUID CRYSTALLINE PHASES WITH SMECTIC ORDERING

L. M. BLINOV, M. I. BARNIK, V. T. LAZAREVA and A. N. TRUFANOV
Organic Intermediates and Dyes Institute, K-1, Moscow, U.S.S.R.

Résumé. — On a étudié les variations avec la fréquence de la tension seuil d'instabilités apparaissant dans les couches de cristal liquide en orientation homogène ou homéotrophe soumises à un champ électrique alternatif. Les cristaux liquides ont des phases nématiques ainsi que des phases smectiques A et leur anisotropie de conductivité électrique change de signe à une température faiblement supérieure à celle de la transition nématique-smectique A. Des données expérimentales sont expliquées à l'aide du mécanisme isotrope et du mécanisme de Carr et Helfrich.

Abstract. — An investigation has been made of the frequency characteristics of the threshold voltage of domain instabilities induced by an electric field in both homogeneously and homeotropically oriented liquid crystal layers. The mesogens had the nematic and smectic A phases and their anisotropy of the electrical conductivity changed its sign at the critical temperature somewhat above the nematic-smectic transition point. All the instabilities observed were of the electrohydrodynamic nature. The experimental results were accounted for in terms of the isotropic and Carr-Helfrich mechanisms.

1. Introduction. — At present, mechanisms of domain instabilities induced by an a.c. electric field have been well studied both theoretically [1-6] and experimentally [7-10] in the case of nematic liquid crystals (NLCs) with positive conductivity anisotropy ($\sigma_o = \sigma_\parallel - \sigma_\perp > 0$; indices $\parallel$ and $\perp$ refer to the director). For homogeneously oriented NLCs with negative or small positive value of dielectric anisotropy ($\epsilon_o = \epsilon_\parallel - \epsilon_\perp$) there are two types of instability. At frequencies below the critical,

$$f < f_c^* = f_c \sqrt{\frac{\epsilon_o}{\epsilon_s}} - 1,$$

where $\xi$ is the Helfrich parameter [7] and $f_c = 4 \pi \sigma / \epsilon_o$, an instability occurs in the form of transverse Williams domains caused by the Carr-Helfrich destabilizing mechanism [1]. In this case, the experimental data [8] are in excellent agreement with the theory [3, 4]. At frequencies above $f_c^*$ an instability arises in the form of transverse pre-chevron domains (just at the threshold) and chevrons (slightly above the threshold). Such an instability is produced by electro-convective phenomena in a non-uniform field which are characteristic of isotropic liquids as well [9, 10]. The same (isotropic) mechanism is also responsible for an instability in the form of fingerprints (at $f < f_c^*$) and Maltese crosses (at $f > f_c$) which occurs in homeotropically oriented NLCs with $\epsilon_o > 0$ [9, 10]. There is no doubt that both the isotropic and Williams domains have an electro-hydrodynamic (EHD) nature. (It should be noted that at certain limitations on the cell thickness, $\epsilon_o$ and $\sigma$ one can also observe the longitudinal static flexo-electric domains induced in NLCs by a d.c. field [11-13]. We are not interested by them here.)

Instabilities appearing in both the smectics A and nematic phases with a certain short-range smectic ordering were studied to a lesser extent. The latter are characterized by a change in sign of the conductivity anisotropy at specific temperature ($T^*$) somewhat above the nematic-smectic transition ($T_{NA}$). Thus, $\sigma_\parallel / \sigma_\perp < 1$ over a temperature range between $T^*$ and $T_{NA}$. Such substances are very convenient when somebody wants to separate contributions of different mechanisms to the observed instability. Indeed, the Carr-Helfrich mechanism does not work at $\epsilon_o < 0$ [6], while, to a first approximation, the isotropic mechanism is not sensitive to the sign of conductivity anisotropy.

Previous investigations of instabilities in NLCs with $\epsilon_o < 0$ were performed only for the particular case of the homogeneous NLC orientation and $\epsilon_o < 0$ [14-16]. The electrohydrodynamic [15, 16] and flexo-electric [16] mechanisms were assumed in order to explain the phenomena observed. In the present paper, a detailed study is made of the field-induced instabilities in both homogeneously and homeotropically oriented NLCs with different values and signs of $\sigma_o$ and $\epsilon_o$. In addition, there have been carried out some investigations of the instabilities in the smectic A phase.
2. Experimental. — The experimental investigations of EHD instability were made on three homologues of the 4-butoxybenzilidene-4'-alkylaniline series,

$$C_4H_5O-\bigcirc-CH=\overset{\wedge}{N}O-\bigcirc-R$$

where \(R=C_6H_5\) (40.4), \(R=C_7H_5\) (40.7) and \(R=C_8H_{17}\) (40.8). The temperature dependences of dielectric permittivity components and the anisotropy of electrical conductivity are shown in figures 1 and 2. All the substances have a negative dielectric anisotropy. The jumps of the curves \(\varepsilon_{\parallel}(T)\) and \(\varepsilon_{\perp}(T)\) correspond to phase transition temperatures, which are given in the following Table:

<table>
<thead>
<tr>
<th>Temperatures, °C</th>
<th>Compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>40.4</td>
</tr>
<tr>
<td>Smectic B (\Rightarrow) Smectic A ((T_{AB})) (\uparrow)</td>
<td>45.2</td>
</tr>
<tr>
<td>Smectic A (\Rightarrow) Nematic ((T_{AN})) (\uparrow)</td>
<td>46</td>
</tr>
<tr>
<td>Sign inversion of (\sigma_s(T^*)) (\uparrow)</td>
<td>51 ± 0.25</td>
</tr>
<tr>
<td>Nematic (\Rightarrow) Isotropic ((T_{NI})) (\uparrow)</td>
<td>75</td>
</tr>
</tbody>
</table>

\(^{\uparrow}\) Data from [17].

Our measurements.

The frequency dependences of the instability threshold were measured on cells of the usual construction, with transparent electrodes of SiO\(_2\). Homogeneous orientation of the liquid crystals was achieved by rubbing the electrodes after their washing in a chrome mixture, homeotropic by thorough mechanical cleaning of the electrodes with subsequent washing in organic solvents. The absence of any molecular reorientation induced by a high-frequency field which could be detected by the phase delay measurements was taken as performance criterion for NLC orientation.

The threshold voltages for formation of domain patterns or onset of motion of solid foreign particles were recorded to an accuracy of \(\pm 5\%\) with the aid of a polarizing microscope.

The value and sign of dielectric anisotropy were controlled by doping the substances with small amounts of either p-cyanophenyl ester of p-heptylbenzoic acid (CEHBA, \(\sigma_s > 0\)) or 2,3-dicyano-4-pentyloxyphenyl ester of p-pentyloxybenzoic acid (DCEPBA, \(\sigma_s < 0\)). The mean conductivity and anisotropy of the conductivity were changed by doping the NLCs with ionic, donor and acceptor impurities [8].

3. Results. — 3.1 HOMOGENEOUS ORIENTATION. —

The frequency dependences of the threshold voltages for instability (\(U_{th}\)) are typical of the three substances under investigation and shown in figure 3 only for substance 40.7. They were measured at temperatures both above and below \(T^*\), that is, for \(\sigma_s > 0\) and \(\sigma_s < 0\). On the curves, two frequency regions can be clearly distinguished.

At high frequencies \(U_{th}\) increases linearly with \(f^{1/2}\) independently of sign of \(\sigma_s\). However its visual appearance depends upon \(\sigma_s\): for \(\sigma_s > 0\) the instability (just at \(U_{th}\)) appears in the form of linear pre-chevron domains with period of about 5 \(\mu\) which are perpendicular to the director (somewhat above \(U_{th}\) the chevrons occurs). At those temperatures, where \(\sigma_s < 0\), linear domains are directed in arbitrary way for various areas and form lattices in the places of their intersections. The domain period is essentially independent of temperature and the threshold voltages are proportional to cell thicknesses (i.e., there is a field threshold).

At low frequencies, there are radically different domains at different temperatures. Curves 2 and 3 in figure 3 correspond to classical Williams domains (the conductance regime at frequencies below the critical). Curves 4 and 5 correspond to the longitudinal domains (|| to the director, see figure 4a) which have been previously observed in [15]. Both the Williams and longitudinal domains can be only observed using light polarized along the director. The temperature behaviour of the threshold of the low-frequency instabilities is shown in figure 5 for various layer thicknesses of substance 40.7. Curves 1-3 and 4
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Fig. 3. - Frequency dependences of the threshold of EHD instability for homogeneously oriented layer of 40.7 \( (\epsilon_n < 0, \ d = 30 \mu) \). Temperature, °C: 84 (1), 78 (2), 70 (3), 62 (4), 57.5 (5).

Fig. 4. - Form of domain patterns \((f = 30 \text{ Hz})\). Homogeneous orientation \((40.7, \epsilon_n < 0, \ d = 12 \mu, \ U = 9 \text{ V})\): \(a\) longitudinal domains \((60 °\text{C})\); \(b\) parquet \((70 °\text{C})\); \(c\) homeotropic orientation \((40.4, \epsilon_n > 0, \ d = 30 \mu, \ U = 35 \text{ V})\) : fingerprint domains at \(T = 50 °\text{C}\).

relate to the longitudinal and Williams domains, respectively. One can see a divergence of \(U_{th}\) for the latters at some temperature \((T')\) near the critical, \(T' \leq T^*\), that is, Williams domains can be also observed at the slightly negative value of \(\epsilon_n\) \(\sigma_{||}/\sigma_{\perp} \approx 0.95\) at \(T'\).

The longitudinal domains are of a EHD character. Solid foreign particles move circularly in the plane perpendicular to domain axes and, at the same time, shift along the domains. The velocity of circular motion increases with increasing voltage. In general, the threshold voltage for longitudinal domains slightly depends upon temperature but sharply increases near the transition to the smectic A phase. The threshold field strength is independent of cell thickness. That is why in thick cells \((d \geq 60 \mu)\) longitudinal domains can be observed only for \(\epsilon_n < 0\) where the threshold for Williams domains diverges. The domain period is comparable with cell thickness, almost independent of voltage and temperature and slightly decreases with increasing frequency. As \(U_{th}\) is proportional to \(d\), the threshold voltage for these domains in thin cells is lower than for Williams domains (curve 1 in Fig. 5). However, longitudinal domains are transformed to parquet type ones when temperature increases above \(T^*\).

The frequency behaviour of the high frequency instability \((f > f_c)\) does not change neither at \(\epsilon_n = 0\) nor at the transition to the smectic A phase though in the latter case, \(U_{th}\) increases sharply (but without any jump) with decreasing temperature.

In the isotropic phase, analogous threshold-frequency curves can be measured with the aid of foreign particles \([10]\). On these curves (see, e.g., curve 1 in Fig. 3), there is a region of square-root dependence at frequencies \(f > f_c\) and a plateau region at \(f < f_c\). One can see in figure 5 that with increasing temperature the curve 5 for high-frequency pre-chevron domains passes smoothly to the square-root region mentioned above, and curve 2 for low-frequency longitudinal domains can be mentally extrapolated to the plateau region.

The threshold voltage for longitudinal domains \((f < f_c)\) increases with increasing mean conductivity as \(U_{th} \sim \sigma^{1/2}\) while that for pre-chevron domains \((f > f_c)\) is independent of \(\sigma\) (see Fig. 6). The variations of \(\sigma_m\) and dielectric anisotropy (over a range of
Fig. 6. — Frequency dependences of threshold voltages, for a homogeneously oriented layer of compound 40.4 (d = 30 μm). Electrical conductivity (ohm cm⁻¹): 8 × 10⁻¹¹ (1), 1.5 × 10⁻¹⁰ (2), 3 × 10⁻¹⁰ (3), 5 × 10⁻¹⁰ (4). Insert: dependence of Uₜ on conductivity (f < 30 Hz).

In the nematic phase, at T⁎ < T < T₅ (curve 2) a domain pattern in the form of Maltese crosses occurs over all the frequency range under investigation and Uₜ ~ f¹/₂. A plateau region where fingerprint domains have to appear [9, 10], locates at lower frequencies.

Further decreasing temperature results in the appearance of two different regions on the Uₜ(f) dependences (curve 3). At high frequencies, where Uₜ ~ f¹/₂, the Maltese crosses are observed over all the temperature interval down to T₅. At low frequencies domains bear a resemblance to fingerprints, see figure 4c. For the same temperatures and the same conductivities the behaviour of the Uₜ(f) curve at 0 < σₐ < 0 and 0 < σₐ < 0 is identical with that for the homogeneously oriented layers having σₐ < 0 and σₐ < 0. It is of importance that the absolute values for the corresponding voltages almost coincide, too. At last, it should be noted that all the features characteristic of longitudinal domains occurring in homogeneously oriented layers (dependences of Uₜ upon σ, σₐ, d and T) also retain for homeotropically oriented layers with σₐ > 0 (at σₐ < 0 the homeotropic layers as well as the homogeneously ones at σₐ > 0 are unstable dielectrically).

As in the case of homogeneous orientation described domain patterns are also observed only over a narrow temperature range of the smectic A phase near T₅. At T ≪ T₅ either novel domain patterns or EHD instabilities assisted by texture changes can be observed only under specific conditions: at small positive dielectric anisotropy or large mean conductivity (σ ≥ 10⁻⁸ ohm⁻¹ cm⁻¹), respectively.

4. Discussion. — In our opinion, all the experimental results can be accounted for using two known mechanisms for the domain formation: the aniso-

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- 1 < εₐ < + 0.1) have no effect on the value Uₜ for longitudinal and pre-chevron domains. The latter measurements have been made on substance 40.4 doped with CEHBA (up to 1.5 %) or DCPEBA (up to 4 %). The dopants shifted transition temperatures of 40.4 less than by 0.5-1 °C. It should be noted that the domain instability was also observed for εₐ = 0 and σₐ = 0 when the dielectric and Helfrich's electrohydrodynamic torques exerted on the director were vanishing.

Similar domain patterns were also observed in the smectic A phase at temperatures just below T₅ (by 0.5-1 °C). At lower temperatures domains disappear but motion of a liquid crystal is still observed. With further decreasing temperature domains appeared and disappeared once more just after the transition to the smectic B phase. Their threshold voltage is higher than that for the homeotropically oriented layers as well as the homogeneously oriented layer of compound 40.4 doped with CEHBA (εₐ > 0, d = 30 μm, σ = 9 × 10⁻¹¹ ohm⁻¹ cm⁻¹ at T = 50 °C). Temperatures, °C: 63 (1), 70 (2), 50 (3).

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FIG. 7. — Frequency dependences of threshold voltages of EHD instability for a homeotropically oriented layer of compound 40.4 doped with CEHBA (εₐ > 0, d = 30 μm, σ = 9 × 10⁻¹¹ ohm⁻¹ cm⁻¹ at T = 50 °C). Temperatures, °C: 63 (1), 70 (2), 50 (3).
tropic Carr-Helfrich model [1] and the isotropic non-injection mechanism [10].

First of all let us discuss the Williams domain regime which takes place for homogeneously oriented layers at frequencies below the critical (curve 4 in Fig. 5). In this case, the temperature dependence of the threshold voltage is due mainly to the temperature dependence of the conductivity anisotropy, though the temperature behaviour of elastic and viscosity coefficients and dielectric permittivities should be also taken into account. Strictly speaking, we have to use the two-dimensional model [4, 5, 8] in order to calculate the threshold voltage. This has not been done here because exact values of all the necessary material parameters were not known. Qualitatively, we can expect from the one-dimensional Carr-Helfrich model [1] that the threshold voltage has to diverge when Helfrich’s parameter,

$$\varepsilon^2 = \left(1 - \frac{\varepsilon_2}{\varepsilon_1} \cdot \frac{\varepsilon_1}{\varepsilon_2} \right) \cdot \left(1 + \frac{\varepsilon_2}{\varepsilon_1} \cdot \frac{\varepsilon_1}{\varepsilon_2} \right),$$

(1)

tends to unity. Here \(\varepsilon_2\) and \(\varepsilon_1 = 1/2(\varepsilon_2 + \varepsilon_2 + \varepsilon_2)\) are viscosities expressed by Leslie's coefficients. From eq. (1) we obtain that \(U_{th} \to \infty\), if

$$\frac{\varepsilon_2}{\varepsilon_1} \to 1 + \frac{\varepsilon_1}{\varepsilon_2} \left(1 + \frac{\varepsilon_1}{\varepsilon_2} \right).$$

(2)

In experiment, \(U_{th} \to \infty\) at \(\varepsilon_2/\varepsilon_1 = 0.95\). Using this value and \(\varepsilon_2 = -0.13, \varepsilon_1 = 3.98\) we can calculate the unusually small ratio \(\eta_1/\alpha_2 \approx 0.5\) and conclude that viscosity \(\alpha_2\) of compound 40.7 is positive. Now, we do not know whether this result remains for the two-dimensional approach or not.

We interpret all the rest instability modes by means of the isotropic mechanism. According to this model [10, 19], an electro-convective instability occurs in any liquid (isotropic or anisotropic) due to the non-uniform distribution of space charge along the field direction, \(dq/dz \sim vE_z\) (\(v\) being the corresponding kinetic coefficient). This model predicts two regions on the threshold-frequency curves: a low-frequency plateau

$$E_{th}^2 \sim \frac{\eta\sigma}{v\varepsilon^2}, f \ll f_c$$

(3)

and a high-frequency root-square region

$$E_{th}^2 \sim \frac{\eta f}{v\varepsilon}, f \gg f_c$$

(4)

where \(\eta, \sigma, \varepsilon\) are the mean viscosity, conductivity and dielectric permittivity. Besides, \(E_{th} = U_{th}/d\) is independent of cell thickness. Our experimental data for both the longitudinal domains and «Maltese crosses» agree qualitatively with eqs. (3) and (4): \(U_{th} \sim \sigma^{1/2}\) at low frequencies and is independent of \(\sigma\) at high frequencies where \(U_{th} \sim f^{1/2}\). The high-frequency threshold increases with decreasing temperature due to the increase in mean viscosity. At the same time, \(U_{th}\) weakly depends upon temperature at low frequencies because of the compensation of the viscosity and conductivity effects. However, near the transition to the smectic A phase \(U_{th}\) increases markedly due to the sharp increase in viscosity caused by the permeation process [18].

The crucial argument for an isotropic formation mechanism of the longitudinal or pre-chevron domains in the case of homogeneous orientation, and the «Maltese crosses» or fingerprints in the case of homeotropic orientation is the absence of any dependence of their thresholds upon anisotropic material parameter as well as an appearance of these instabilities at \(\varepsilon_2 = 0\) and \(\varepsilon_1 = 0\). Of course, the anisotropy of electrical conductivity can modify the space charge distribution, and the anisotropy of viscosity as well as liquid crystal orientation can affect the topology of flows responsible for domain patterns observed in experiments. Thus, it seems to be very promising to consider theoretically the problem of the isotropic instability with account taken of various anisotropic parameters of liquid crystals.

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References
