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To cite this version:

R. Ribotta, G. Durand. HIGH FREQUENCY ELECTROHYDRODYNAMICAL INSTABILITIES IN NEMATIC LIQUID CRYSTALS. Journal de Physique Colloques, 1979, 40 (C3), pp.C3-334-C3-337. <10.1051/jphyscol:1979365>. <jpa-00218761>

HAL Id: jpa-00218761
https://hal.archives-ouvertes.fr/jpa-00218761
Submitted on 1 Jan 1979
HIGH FREQUENCY ELECTROHYDRODYNAMICAL INSTABILITIES
IN NEMATIC LIQUID CRYSTALS

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Résumé. — Nous avons mesuré les seuils des instabilités électrohydrodynamiques (EHD) apparaissant dans la texture planaire du cristal liquide nématique MBBA (p-méthoxybenzilidène-p-n-butyl-aniline), au-dessus de la fréquence de relaxation des charges. Nous observons deux instabilités distinctes : un mouvement convectif lent d’écoulement, rendu visible par des mouvements de poussière, sur des distances de l’ordre de l’épaisseur de l’échantillon ; le réseau de lignes serrées de l’instabilité diélectrique, de longueur d’onde de quelques microns, donnant à plus forte excitation les chevrons. À température fixe, l’instabilité convective à un seuil inférieur à celui des lignes diélectriques. Ces deux instabilités restent découpées probablement à cause de leur dépendance spatio-temporelle très différente. À fréquence fixe, la variation en température montre pour le mode convectif une continuité, à $T_\text{c}$ (transition nématique-isotrope), avec une instabilité isotrope de type Felici. À l’inverse, le seuil des lignes diélectriques diverge à $T_\text{c}$, comme le prévoit le mécanisme de Carr-Helfrich. Nos résultats confirment la validité du modèle de Carr-Helfrich pour l’instabilité EHD de régime diélectrique. La récente affirmation de Barnik et al. que l’instabilité diélectrique de haute fréquence s’expliquait par un modèle de type Felici isotrope pourrait provenir d’une confusion entre les deux types d’instabilités.

Abstract. — We have measured the thresholds of the electrohydrodynamic (EHD) instabilities appearing in the planar texture of the nematic liquid crystal MBBA (p-methoxybenzilidene-p-n-butyl-aniline) above the charge relaxation frequency. We observe two distinct instabilities, a slow convective hydrodynamic motion (made visible by dust motion) over distances comparable to the sample thickness, and the much shorter wavelength dielectric stripes (which give rise at higher voltage to the chevrons). At fixed temperature, the convective instability has a lower threshold than the dielectric stripes. These two instabilities are not coupled probably because of their very different spatial and time dependence. For fixed frequency, the temperature dependence of the convective threshold shows a continuity at $T_\text{c}$ (nematic to isotropic transition), with an isotropic Felici-like instability. On the contrary, the dielectric threshold diverges below $T_\text{c}$ as expected in the Carr-Helfrich mechanism. Our results confirm the Carr-Helfrich mechanism, to explain the dielectric EHD instability. The claim by Barnik et al. that the high frequency EHD dielectric instability is explained by the Felici isotropic mechanism might result from a confusion between the two kinds of instabilities.

Electrohydrodynamical (EHD) instabilities in nematic liquid crystals have arisen in the past a great interest, because of their applications to displays [1]. Following an idea by Carr [2], Helfrich [3] suggested a possible mechanism for DC excitation, extended by the Orsay Group [4] to the AC regime. This mechanism was based on a parametric coupling between space charges and bend distortion through the application of an electric field. It explained, in particular, the so-called dielectric regime, for an excitation frequency larger than the charge relaxation frequency (of the order of 10-100 Hz in a typical nematic liquid crystal). This mode was understood as a bend oscillation synchronous to the applied electric field oscillation, around a quasi-static space charge distribution. The wave-vector of this mode was predicted, and found, much larger than the inverse thickness of the sample (on the contrary of the low frequency regime resulting in the so-called Williams domains which appear with a spatial period comparable to the sample thickness). Just above threshold, the thin periodic lines (period ~ a few microns) which are identified with the dielectric oscillations undergo a distortion and produce a typical chevron pattern. A lot of work has been devoted to these instabilities confirming the validity of the Carr-Helfrich-Orsay (CHO) model. Let us mention, for instance, the magnetic field dependence of the spatial period of the dielectric oscillations [5], and the observation of pretransitional bend oscillations by Rayleigh scattering [6].

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More recently, a Russian Group [7] underwent a systematic study of threshold values associated with high frequency EHD instabilities. A temperature dependence of the measured threshold has been published which showed no discontinuity at the transition temperature \( T_c \) (from the nematic to the isotropic phase) with EHD instabilities observed in the isotropic phase. These last instabilities can be explained by one of the models described by Felici [8a] and Atten [8b] for isotropic liquids. The conclusion of the Russian Group is that the high frequency dielectric regime was not due to the CHO mechanism, but was the particular appearance, in the nematic phase, of a Felici-like instability characteristic of isotropic liquids.

The purpose of the present paper is to resume threshold experiments for the EHD instabilities in the dielectric regime, to ascertain which model, CHO or isotropic Felici, explains really the onset of high frequency instabilities.

1. The experimental set-up. — We have used the standard geometry which is now classical for the observation of this kind of instabilities. We use a planar nematic sample of MBBA (methoxybenzilidene butyl aniline), with a \( T_c \) of the order of 43 °C. This compound is not very pure but has the same \( T_c \) as the MBBA used in the Russian work [7]. The planar alignment is obtained by rubbing tin oxide coated glass plates. The thickness of the sample can be adjusted between 20 and 200 \( \mu \)m. The sample is placed inside a temperature controled oven (±0.2 °C) and observed under a polarizing microscope. An AC generator applies a variable amplitude (V) and frequency (\( \omega \)) sine-wave voltage across the nematic sample. Some dust particles in the field of observation can show by their individual or correlated motion the existence of a field induced flow in the sample.

2. Experimental results. — We first chose a rather thin sample (\( d = 26 \mu \)m) to reproduce the observations of reference [7]. At low frequency, we observe regular Williams domains. The cut-off frequency, close to the charge relaxation frequency \( f_c \), is found at about 40 Hz. Due to sample aging, \( f_c \) can increase up to 130 Hz after one month. Increasing the frequency, we do observe the threshold dependence of the dielectric oscillations proportional to \( \omega^{1/2} \) (see Fig. 1, dots). This threshold is very sharp and can be determined with a one per cent accuracy. The interesting feature is that, below this threshold, we can see already some flow in the nematic, through the displacement of dust particles. Increasing the applied voltage from zero, for a given frequency, we first observe a slow erratic and uncorrelated motion of individual dust particles, always along the nematic director. For a higher voltage, we observe a correlated slow motion of almost all the dust particles with motion across the director. This convective flow seems to proceed on distances of the order of 20 to 40 \( \mu \)m measured along the electrodes. We have plotted the threshold for this large scale motion of dust particles (crosses on Fig. 1). Note that the two thresholds (dielectric stripes and convective flow) are well distinct in the low frequency range but that, beyond 500 Hz, these two thresholds coincide. We reproduce now the same experiment with a thicker sample, (width \( d = 104 \mu \)m) and we find again two distinct threshold, the smaller for a slow correlated flow of dust particles, the larger for the EHD dielectric stripes. Our results are plotted on figure 2. The difference with the case of the thin sample is that now, within the amplitude capability of our AC generator (500 V rms max), the two thresholds remain distinct, especially at frequencies above 500 Hz where the two thresholds coincided for the thin sample. Because of the larger sample thickness, using the short field of focus of the microscope, we can really ascertain that the flow of dust particles is a real convective flow, on a typical horizontal scale of 50 to 100 \( \mu \), and a vertical scale a little smaller than the sample thickness.
Fixing now the frequency to 200 Hz, we have measured the temperature dependence of the two thresholds (dielectric stripes and convective flow) for the two different thickness samples. Figure 3 shows the behaviour of the thin (26 μm) sample. Close to room temperature, the two thresholds for the dielectric stripes and the convective dust flow are very close to one another. As T is increased, these two thresholds separate from each other. The convective threshold decreases; this probably follows the temperature decrease of the mean viscosity. As observed in reference [7], we do find the same kind of turbulent flow in the isotropic phase of MBBA, with a continuous threshold variation across $T_c$. On the opposite, the dielectric stripes threshold increases with increasing temperature and appears to diverge close to $T_c$. Above $T_c$, absolutely no isotropic instability threshold is observed which could be continuous at $T_c$ with the dielectric stripes threshold.

The same experiment is done using a thick sample ($d = 104 μm$). At room temperature, we do observe the two different thresholds, the lower for the convective motion, the larger for the dielectric stripes. These two thresholds are now well separated, by a factor of 2 rather than by the few percent typical of the thin sample (see Fig. 4). Here again, we do observe a continuity at $T_e$ for the convective instability threshold, and a divergence below and close to $T_e$ for the dielectric stripes threshold.

3. Discussion. — The divergence of the dielectric stripes threshold is easy to understand in the frame of the CH0 model. Let us call $η$ the bend viscosity and $\varepsilon_a$ the anisotropy of the dielectric constant; $\varepsilon_a$ is the difference between the two eigen-values of the dielectric tensor $\varepsilon_{ij}$ along the director and $\varepsilon_{ij}$ across the director. The anisotropy $\varepsilon_a = \varepsilon_3 - \varepsilon_1$ is negative for MBBA. Increasing the temperature, $|\varepsilon_a|$ decreases and vanishes at $T_c$. On the other hand, $η$ is regular at $T_c$. The threshold for dielectric stripes corresponds to an electric field such that the dielectric contribution to the bend damping rate $\frac{1}{2} \frac{|\varepsilon_a| E^2}{4 \pi \eta}$ is of the order of the frequency $\omega$ (see Ref. [1]). Close to $T_e$, $|\varepsilon_a|$ vanishes. As $η$ is regular, one expects a divergence of the threshold field $E$ as $|\varepsilon_a|^{-1/2}$. We have plotted on figure 5 the temperature dependence of the inverse squared thresholds $E^{-2}$ versus $ΔT = T_c - T$. The voltage thresholds for the two different thickness samples do not follow exactly a thickness dependence law as a true field threshold would. We have corrected for that variation arbitrarily by a scale adjustment. We obtain now one single curve to represent the temperature dependence of the dielectric stripes thresholds for the two samples. On the same graph, at the same arbitrary scale is plotted the temperature dependence
of $\epsilon_\alpha$, known from an independent work [9]. Within 10%, the two curves are identical. The systematic departure observed could be accounted for by the neglected temperature dependence of the viscosity and the elastic constants, which should have been taken into account to explain at least the regular variation of threshold far from $T_c$. On the other hand, the continuity of the convective motion threshold at $T_c$ is well a proof, as noted by the Russian Group, that the mechanism leading to this instability does not depend on the nematic order. It could be either the Felici [8a] mechanism or any other such as unipolar injection already in low conductivity [8b] materials proposed for isotropic liquids. Up to now, the proper mechanism responsible for that instability has not been found.

One can now wonder why the two instabilities, the dielectric stripes and the convective motion, do not couple altogether, since they present in general two different thresholds. We note that these two thresholds are well distinct in the thin sample, in the low frequency range. They remain distinct, whatever may be the frequency, in the large thickness sample. A possible explanation of the weak coupling of these two modes could be due to their very different spatial dependence, the dielectric stripes appearing on a scale of a few microns, compared to the sample thickness 26 or 104 $\mu$m that give the scale of the convective vortices. Another reason is probably the very different time scale associated with the flow in these two regimes. The dielectric stripes instability is accompanied by a forced high frequency alternative flow at the excitation frequency $\omega_0$ whereas the convective instability appears as a DC flow. The situation is completely different in the DC limit of the low frequency (conduction) regime. The CHO and the isotropic mechanism are expected to have a maximum coupling, since they lead independently to vortices of the same size and frequency. This coupling could explain the apparent continuity at $T_c$ between Williams domains in the nematic phase and convective flow in the isotropic case [10].

4. Conclusion. — In the frequency regime of the EHD instabilities in nematic liquid crystals, we have observed two different unstable modes. One is a convective motion leading to vortices of a size comparable to the sample thickness. As previously reported [7], this mode is continuous at the nematic to isotropic phase transition $T_c$. It is reminiscent of the EHD instabilities described by Felici for isotropic dielectric fluids. The other mode is the well-known dielectric stripes instability which leads to the chevrons texture. This last instability is characteristic of the nematic phase. Its threshold diverges at $T_c$, as predicted by the Carr-Helfrich-Orsay model. In general, these two modes, which result from two different mechanisms, are well distinguishable by careful observation. In some restricted cases (thin samples, high frequency, low temperature, no analysis of the spatial dependence) these two unstable modes could be confused.

We suggest that such a confusion may have occurred in reference [7], which incorrectly concluded that the high frequency dielectric instability had the isotropic character. It would be interesting to study further the isotropic convective flow in the low frequency conduction regime to explain its possible role in the onset of the often described but not really understood dynamic scattering mode.

References

[1] For a review of these instabilities and general references, see Durand, G., in Les Houches summer School (Gordon and Breach) 1973, p. 402-440.
[7] BARNIK, M. I., BLINOV, L. M., GREBENKIN, M. F., TRUPA-