INSTABILITIES IN ELECTRIC FIELDS OF NEMATIC LIQUID CRYSTALS WITH POSITIVE DIELECTRIC ANISOTROPY: DOMAINS, LOOP DOMAINS AND REORIENTATION

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Instabilities in electric fields are reported for p,p'-di-*n*-butyl azoxybenzene (DIBAB), a new room temperature nematic with a positive dielectric anisotropy. The threshold voltage is found to be almost independent of the frequency up to 100 kHz. However, unlike in nematics with negative dielectric anisotropy usually no normal domains are observed. The instabilities take the form of a reorientation, while loop domains are observed as a transient effect. These loop domains indicate that the instability is not purely dielectric in origin.

If a thin layer of a nematic liquid crystal in the planar texture is subjected to a dc or ac electric field, instability occurs above a threshold voltage. Attention has been given mainly to nematics with negative dielectric anisotropy ($\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp < 0$) where the well-known cellular flow patterns (Williams domains) are observed [1–4]. In this letter we give results for a new room temperature nematic liquid crystal with positive dielectric anisotropy.

In dc fields a net injection of charge carriers into an isotropic dielectric liquid leads to space charges. As in the classical Bénard problem these space charges lead to cellular flow [5]. This mechanism is also present in the nematic phase. This explanation is consistent with the observation that in dc fields the cellular flow persists in the isotropic phase [6]. As injection disappears above a few Hz the instabilities in ac fields cannot be explained in this way. In a nematic, however, space charges can also arise due to the anisotropic conductivity ($\sigma_\parallel > \sigma_\perp$) [7]. For nematics with $\Delta\epsilon < 0$ this is clearly the relevant mechanism for the flow in ac fields, in agreement with the observation that no cellular flow is found in the isotropic phase [4].

Helfrich has made detailed calculations [8] of the torques on a nematic liquid crystal in an electric field, which have recently been extended to ac fields [9]. In this theory spatial variations of the molecular orientation (as given by the director $\boldsymbol{n}$) are considered for an infinite nematic. $\boldsymbol{n}$ is supposed to vary in the X direction only (see fig. 1a). The original direction is thought to be stabilized by a magnetic field H. The threshold field for static distortions is given by:

$$\overline{E_c^2} = \frac{1+\omega^2\tau^2}{\Delta\epsilon\,\omega^2\tau^2+\Theta_H}\,\frac{4\pi\epsilon_\parallel}{\epsilon_\perp}\left(k_{33}\frac{\pi^2}{d^2}+\Delta\chi H^2\right). \tag{1}$$

τ is the dielectric relaxation time given by $\epsilon/4\pi\sigma$, k_{33} is the elastic constant for bend, Θ_H is a parameter depending on the dielectric constant, the conductivity and on viscosity coefficients, and therefore characteristic of the particular nematic compound. In all practical cases Θ_H is positive and of the order of 1.

For $\Delta\epsilon < 0$ eq. (1) is only valid for frequencies up to τ^{-1}: the "conduction" regime [4, 10]. In this region the space charges oscillate with the frequency

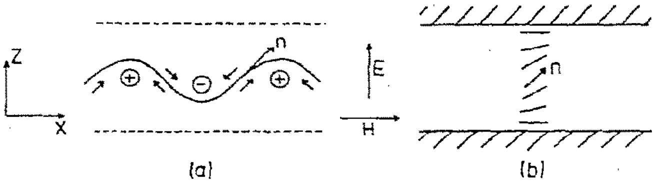


Fig. 1. (a) Carr–Helfrich-type instability: $\boldsymbol{n} = n(x)$; (b) Freedericks-type instability: $\boldsymbol{n} = n(z)$.

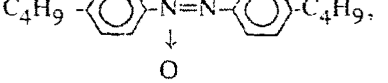
while the molecular distortions are static. Above the critical frequency τ^{-1} the approximations underlying eq. (1) break down. We enter the "dielectric" regime in which the space charges become static and the molecular distortions oscillate. For $\Delta\epsilon > 0$, on the other hand, the denominator in eq. (1) is always positive: the conduction regime extends to high frequencies.

Instead of an electrohydrodynamic instability, one could expect for a nematic with positive $\Delta\epsilon$ dielectric alignment of the molecules parallel to the field. Then $\boldsymbol{n}$ is supposed to depend only on the Z direction (fig. 1b). Analogous to Saupe's work [11] one finds:

$$\overline{(E_c')^2} = \frac{4\pi}{\Delta\epsilon}\left(k_{11}\frac{\pi^2}{d^2} + \Delta\chi H^2\right), \qquad (2)$$

where the initial orientation is again stabilized by a magnetic field $H \perp E$, and k_{11} is the elastic constant for splay. One might wonder whether the theoretical distinction between E_c and E_c' is real; a full two-dimensional theory ($\boldsymbol{n}$ dependent on X and Z) would probably give only one threshold depending on both k_{11} and k_{33}.

Measurements on compounds with positive $\Delta\epsilon$ have been reported only for p,p'-di-*n*-heptyloxy azobenzene [12] which might not be a typical example because of the very small value of $\Delta\epsilon$ (≈ 0.03). Recently p,p'-di-*n*-alkyl substituted azo- and azoxybenzenes have been synthetized by van der Veen et al. [13]. In our experiments we used p,p'-di-*n*-butyl azoxybenzene (DIBAB):



which has a nematic range from 14° to 28 °C. Furthermore $\epsilon_{\parallel} \approx 4.2$ and $\epsilon_{\perp} \approx 4.0$. Similar results as for DIBAB were found for some p,p'-di-*n*-alkyl azobenzenes that have $\Delta\epsilon \approx 0.4$.

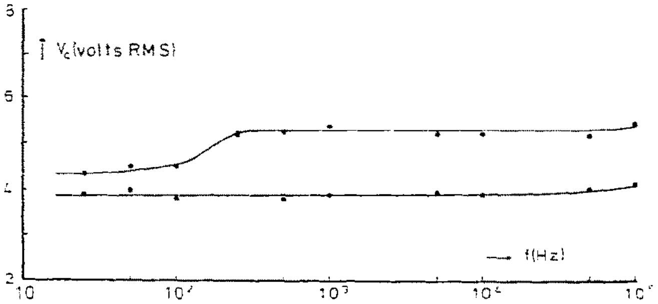


Fig. 2. Threshold for instability in a 50 μm sample; lower curve: pure DIBAB ($\rho \approx 5 \times 10^{10}$ Ωcm); upper curve: doped DIBAB ($\rho \approx 5 \times 10^{9}$ Ωcm).

DC and ac electric fields were applied to DIBAB in the usual planar configuration with the molecules aligned parallel to the rubbed walls. No normal domains are observed in contrast with compounds with $\Delta\epsilon < 0$, but at 4–6 volt a reorientation is found. The molecules switch from their original orientation to a situation more parallel to the field. This can be

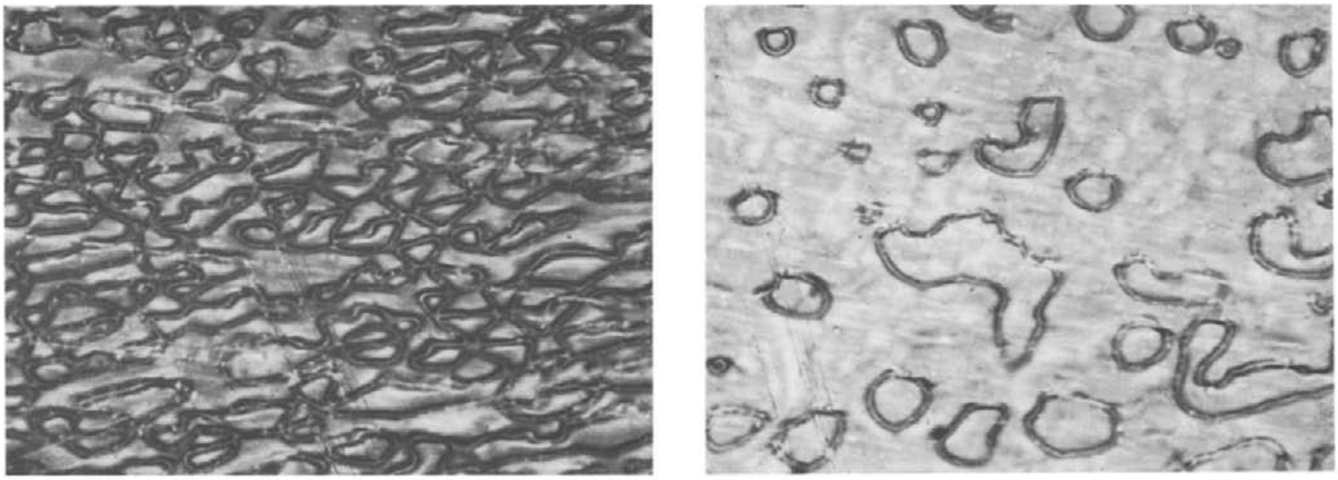


Fig. 3. Loop domains on increasing the voltage (50 Hz) in the doped DIBAB sample (magnification ≈ 80×).

concluded from the change in capacitance of the sample [14]. When passing the threshold a colour change is observed between crossed polarizers. Furthermore, domains are generally seen as a transient effect: loop domains [15] (only for increasing voltage). In order to obtain accurate threshold values the sample was placed between crossed polarizers in a parallel light beam from a He–Ne laser. By recording the transmitted light as a function of the slowly increasing voltage over the sample, the threshold voltage could be determined within 0.2 volt. The results are given in fig. 2 up to a frequency of 100 kHz. The threshold voltage was found to be independent of the thickness of the nematic layer, in agreement with both eqs. (1) and (2).

The experiments were repeated with a DIBAB sample with a conductivity increased 10-fold (obtained by doping). At higher frequencies the threshold is again constant (fig. 2). However, now there is a drop in the threshold voltage below 200 Hz. In this low-frequency region normal stable domains are observed between 4.5 and 5.5 volt. When the voltage is further increased these domains change into loop domains (of the same form as observed at the threshold at higher frequencies) and disappear (see fig. 3). This low-frequency region seems rather similar to the results of Gruler and Meier [12].

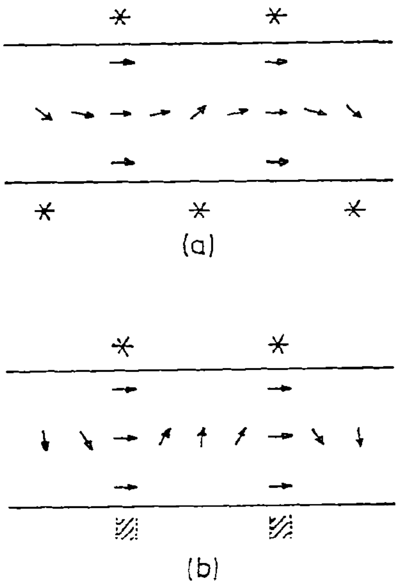


Fig. 4. Orientation in the bulk of the nematic for domains (a) and on increasing the voltage (b). The asterisks indicate bright lines, the broad dark lines are shaded.

The occurrence of loop domains as a transient effect at all frequencies indicates that the origin of the instability is not purely dielectric. Hence eq. (1) seems to be more relevant than eq. (2). Moreover the decrease in threshold in the low-frequency region for the doped sample is one of the predicted possibilities with eq. (1) [9]. The constancy of V_c of the pure sample is somewhat surprising as this means that $\Delta\epsilon \approx \Theta_{II}$. Moreover, it is not clear why the doped sample has a somewhat higher threshold, as the high-frequency limit of eq. (1) is independent of the conductivity. Probably a full two-dimensional theory would clarify these points.

The exact nature of the loop domains can be established by starting with a doped sample with stable Williams domains at, e.g., 5 volt, 50 Hz (crossed polarizers). The spatial distribution of $\boldsymbol{n}$ in the bulk of the slab is given in fig. 4a according to Penz [3]. We focus on the bright lines (real images) *below* the nematic layer. When the voltage is gradually increased the bright lines broaden to bright areas (the focal length increases) with dark lines in between. The movement of dust particles indicates that there is still hydrodynamic flow. At a later stage the dark lines close and form loops (fig. 3). The cellular flow gradually disappears and it is also not necessary to focus below the slab in order to see the loops. On focussing above the slab we notice that the upper series of bright lines is still present right in the middle of the dark lines (fig. 3). The approximate change in orientation of n is given in fig. 4b. The loops are regions where n is still parallel to the walls in surroundings that have started to line up with the external field. The original focussing power that leads to the upper set of bright lines is still present.

In conclusion, we emphasize that the instabilities in DIBAB are not purely dielectric in origin, but are associated with hydrodynamic flow, just as for nematics with negative $\Delta\epsilon$. The difference between the two cases comes in *above* the threshold, where the flow disappears in the case $\Delta\epsilon > 0$ and becomes stronger (turbulence) for nematics with $\Delta\epsilon < 0$. This difference cannot be explained by the existing theories that derive stability conditions only. A discussion is necessary of the situation above the threshold of instability, which will be given elsewhere [16].

Finally we mention that at high voltages (≈ 100 volt rms) some other periodic patterns are often observed in the samples. Similar effects are found in homeotropic samples of DIBAB. However, the reproducibility and interpretation of these high-voltage instabilities is difficult. The results will be reported at a later date.

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