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A new type of electrohydrodynamic instability in nematic liquid crystals with positive dielectric anisotropy

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A new type of electrohydrodynamic instability originally reported in nematic liquid crystal mixtures with positive dielectric anisotropy and as moderately thick samples is further studied. The ability of homogeneously aligned nematics with positive dielectric anisotropy, in the presence of a magnetic field, to exhibit Williams domains as a threshold effect is numerically investigated. The variation of the threshold voltage for domain formation and dielectric alignment with dielectric anisotropy is calculated theoretically and compared with the experimental results as moderately thick and thin samples.

1. Introduction

The existence of electrohydrodynamic (EHD) instability in nematic liquid crystals with positive dielectric anisotropy has been extensively investigated [1–18]. The ability of a nematic liquid crystal layer with positive dielectric anisotropy to exhibit Williams domains [19] as a threshold effect depends on the conductivity, dielectric anisotropies and other experimental conditions as discussed earlier [1, 2]. The ability of homogeneously aligned, positive $\Delta\varepsilon$ nematics, under the influence of a stabilizing magnetic field, to exhibit domains is greater as compared to the configuration where there is no stabilizing magnetic field. It has already been reported [18] that the thickness of the sample will also play a role in the formation of Williams domains as a threshold effect. The stability of the domains is also related [1] to the magnitude of the dielectric anisotropy in positive $\Delta\varepsilon$ nematics.

The present work was to investigate the effect of the stabilizing magnetic field on the domain formation in nematic liquid crystal mixtures with positive dielectric anisotropy. These investigations have been carried out using moderately thick and thin samples for comparison. The variation of threshold voltage for domain formation and dielectric alignment for positive $\Delta\varepsilon$ nematics is theoretically calculated and compared with experimental values for moderately thick and thin samples. A new electrohydrodynamic instability in the form of a grid pattern in high positive $\Delta\varepsilon$ nematics reported earlier [1] is further studied.

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Mixtures of MBBA (4-methoxybenzylidene-4′-*n*-butylaniline) and EBCA (4-ethoxybenzylidene-4′-cyanoaniline) have been used to obtain different values of positive dielectric anisotropy as reported [1] in the earlier publication.

2. Experimental

Dielectric constant measurements are made at a frequency of the order of the applied electric field, since this frequency is of primary importance in the orientation of the molecular axes and consequent instabilities. Due to experimental limitations, dielectric constant measurements have been carried out at 100 kHz using a crystal-controlled oscillator as mentioned previously [1, 2].

The MBBA and EBCA samples were prepared in our laboratory using standard procedures. MBBA was purified by fractional distillation under reduced pressure and EBCA was recrystallized twice before use. The concentration of EBCA in MBBA used in this work was very low, so that the viscosity of the mixture would not be much different from that of MBBA. The AC conductivities of the mixtures were of the order of $10^{-10}\,\Omega^{-1}\,\mathrm{cm}^{-1}$. The conductivity ratio $\sigma_{\parallel}/\sigma_{\perp}$ was 1·3 for MBBA, measured at 80 Hz. This shows a slight increase as the EBCA concentration increases.

A uniform and moderate sample thickness of 250 μm was used for the present work. A sample thickness of 40 μm was employed for the experimental investigation of thin samples. Homogeneous alignment in thin samples was obtained by the rubbing technique. The threshold voltage for thin samples was measured by using a photomultiplier and the optical observations were made microscopically.

3. Results and discussions

The results obtained in studying the effect of the dielectric anisotropy and the magnetic field on the domain formation and dielectric alignment, in addition to the variation of threshold voltages with dielectric anisotropy for positive $\Delta\varepsilon$ nematics are now discussed.

3.1. *The effect of dielectric anisotropy*

Figure 1 shows the variation of threshold voltage with frequency at different values of the dielectric anisotropy for MBBA–EBCA mixtures with $H \perp E$; sample thickness $= 250\,\mu$m, $H = 5{\cdot}7$ kG. The curved part of each curve corresponds to the domain formation and the linear part of the curve to the Fréedericksz deformation [20]. It is evident from the figure that the domain part of the curves is relatively insensitive to the variation of $\Delta\varepsilon$ and the frequency limit of this part, at which the domain and dielectric alignment thresholds become equal, depends strongly on $\Delta\varepsilon$.

At $\Delta\varepsilon = +0{\cdot}1$, stable domains, followed by reorientation and at $\Delta\varepsilon = +0{\cdot}4$, unstable domains with reorientation have been observed. At $\Delta\varepsilon = 0{\cdot}65$, a grid pattern with reorientation and at $\Delta\varepsilon = +1{\cdot}28$ only pure reorientation have been observed. The dielectric constant ratio $\varepsilon_{\parallel}/\varepsilon_{\perp} = 1{\cdot}2$ agrees with the theoretical value of Penz [12] above which there is only Fréedericksz deformation.

3.2. *The effect of a magnetic field*

Figure 2 shows the variation of threshold voltage with frequency at different magnetic field strengths for $H \perp E$, $\Delta\varepsilon = +0{\cdot}4$, sample thickness 250 μm. The graph indicates that the stabilizing magnetic field may cause a positive dielectric nematic, that normally does not form Williams domains, to do so when the magnetic field reaches a certain value. This is due to the competing effects of dielectric and conduction torques

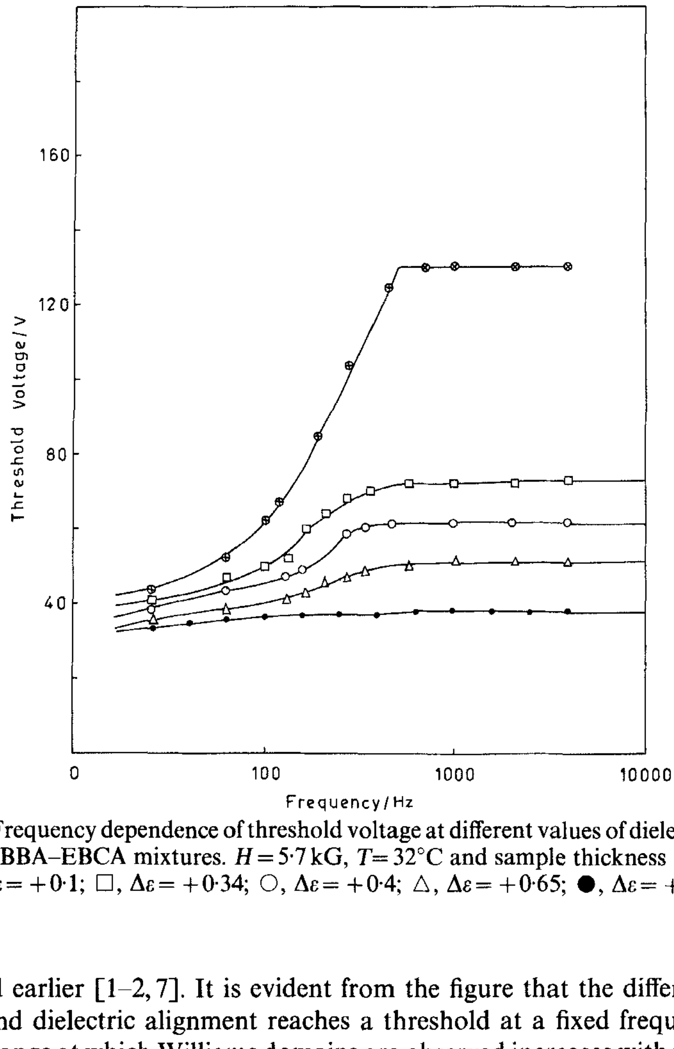

Figure 1. Frequency dependence of threshold voltage at different values of dielectric anisotropy for MBBA–EBCA mixtures. $H=5{\cdot}7$ kG, $T=32$°C and sample thickness $=250\,\mu$m, $H_{\perp}E$. ⊗, $\Delta\varepsilon=+0{\cdot}1$; □, $\Delta\varepsilon=+0{\cdot}34$; ○, $\Delta\varepsilon=+0{\cdot}4$; △, $\Delta\varepsilon=+0{\cdot}65$; ●, $\Delta\varepsilon=+1{\cdot}28$.

as specified earlier [1–2, 7]. It is evident from the figure that the difference between domains and dielectric alignment reaches a threshold at a fixed frequency, and the frequency range at which Williams domains are observed increases with the strength of the magnetic field. There are three types of positive $\Delta\varepsilon$ nematic with the planar configuration obtained by a magnetic field as reported in [16]; the weakly positive ($\Delta\varepsilon < +0{\cdot}5$) nematics in which domain formation normally occurs, moderately positive ($+0{\cdot}5 < \Delta\varepsilon < 1{\cdot}6$) nematics in which domain formation does not normally occur, but domains are formed under the action of a stabilizing magnetic field, and strongly positive ($\Delta\varepsilon > +1{\cdot}6$)—nematics for which domain formation does not occur whatever the strength of the applied magnetic field. Our experimental results regarding the stabilizing magnetic field in positive $\Delta\varepsilon$ nematics are in full agreement with the results reported by Zenginoglou *et al.* [16].

3.3. *Variation of threshold voltage with* $\Delta\varepsilon$

Figure 3 shows the variation of threshold voltage for domain formation (V_{th}) and the dielectric alignment (V_F) with dielectric anisotropy for $H_{\perp}E$ for MBBA–EBCA mixtures; sample thickness is 250 μm. Line 1 represents variation of V_{th} with dielectric anisotropy at 40 Hz. Curve 2 illustrates the variation of V_F at 3 kHz with positive $\Delta\varepsilon$. Curves 3 and 4 show the variation of threshold voltage for domain formation and

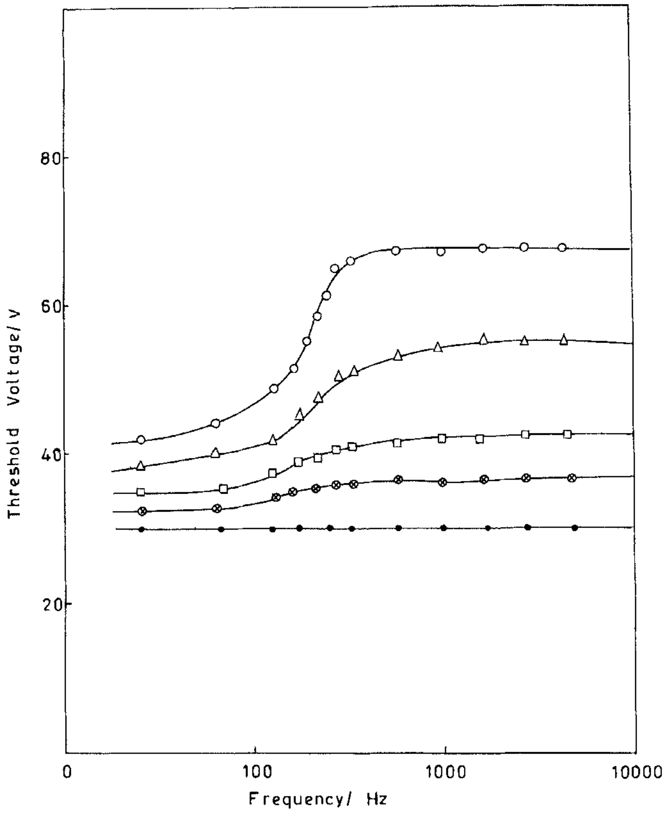

Figure 2. Frequency dependence of threshold voltage at different values of magnetic field strength for MBBA–EBCA mixtures. $\Delta\varepsilon = +0{\cdot}4$, $T = 32°C$, sample thickness $= 250\,\mu m$. ○, $H = 5{\cdot}7$ kG; △, $H = 5{\cdot}15$ kG; □, $H = 4{\cdot}55$ kG; ⊗, $H = 3{\cdot}9$ kG; ●, $H = 0$ kG.

dielectric alignment calculated numerically using the following equations. The threshold field [17] for domain formation is given by the equation

$$E^2 = 4\pi\frac{\varepsilon_{\|}}{\varepsilon_{\perp}}\left(\chi_a H^2 + K_{33}\frac{\pi^2}{d^2}\right)\frac{1+\omega^2\tau^2}{\Delta\varepsilon\omega^2\tau^2 + \theta_H}$$

$$\theta_H = \frac{\varepsilon_{\|}}{\sigma_{\|}}\left[\sigma_a\delta + \frac{\sigma_{\perp}}{\varepsilon_{\perp}}\Delta\varepsilon(1-\delta)\right],$$

where τ is the dielectric relaxation time given by $\varepsilon_{\|}/4\pi\sigma_{\|}$, K_{33} is the bend elastic constant and δ is a dimensionless ratio involving coefficients, the value of $\delta = 0{\cdot}7$ for MBBA, as reported in [17]. The well-known formula [9] for the threshold voltage for the Fréedericksz deformation or dielectric alignment is

$$V_F = 2\pi(\pi K_{11} + \chi_a H^2 d^2)^{1/2}/\Delta\varepsilon^{1/2}.$$

For theoretical calculations, some of the physical constants have been taken from earlier publications on MBBA [13, 18, 21] and the dielectric constants of MBBA–EBCA mixtures were taken from the data by Meyerhofer [22]. The conductivity of MBBA ($\sigma_{\perp} \geqslant 5\times10^{-11}\,\Omega^{-1}\,cm^{-1}$, $\sigma_{\|}/\sigma_{\perp} \approx 0{\cdot}73$) has also been taken from earlier data

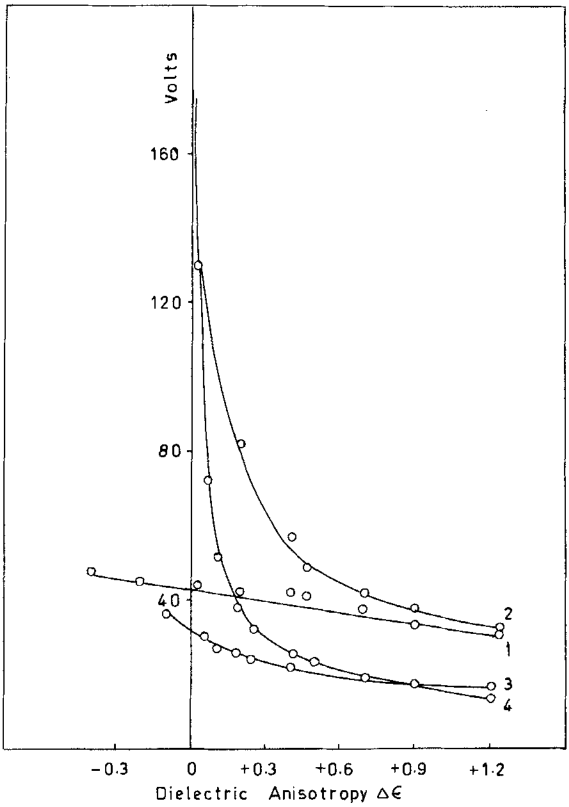

Figure 3. The threshold voltage for domain formation and dielectric alignment for MBBA–EBCA mixtures as a function of the dielectric anisotropy $\Delta\varepsilon$. $H = 5{\cdot}7$ kG, $T = 32$°C and sample thickness = 250 μm.

[18]. The conductivities of mixtures with low concentrations of EBCA in MBBA have been taken as constant for theoretical calculations.

The stability of domains related to various values of the dielectric anisotropies has been explained in our earlier publication [1, 2] in detail. Curves 3 and 4 in figure 3 show that the EHD instability does not exist beyond around $+0{\cdot}9$, but experimentally it is around $+1{\cdot}2$ for a 250 μm thick sample. This may be due to the fact that certain parameters like the conductivities and elastic constants have been taken as constant at different concentrations of EBCA in MBBA for the theoretical calculations, since relevant data are not available in the literature.

Figure 4 illustrates the variation of threshold voltage for domain formation and dielectric alignment at a sample thickness of 40 μm, with an initial homogeneous alignment for the same MBBA–EBCA mixtures. Line 1 and curve 2 are theoretically calculated threshold voltages for domain formation and dielectric alignment using equations reported by Barnik *et al.* [13]. Line 3 and curve 4 are the experimental values for the thresholds for domain formation and dielectric alignment. It is evident from the graph that the calculated positive $\Delta\varepsilon$ beyond which EHD instability no longer exists is greater compared to the experimental value. This may be due to the reasons cited above for moderately thick samples.

The new type of EHD instability, in the form of a grid pattern, in moderately thick samples and reported earlier [1, 2] (see figure 5), has been studied further. It is

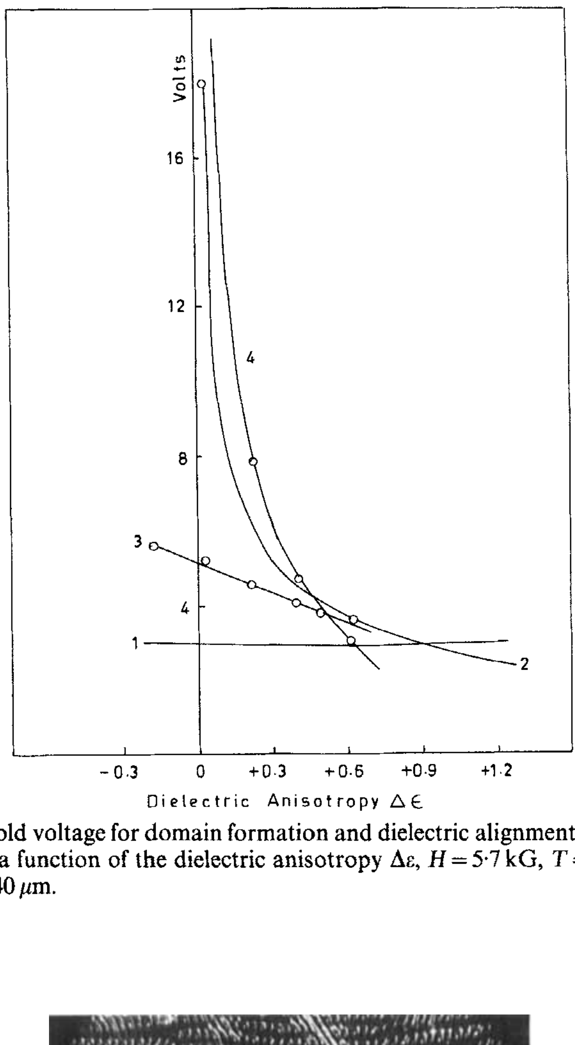

Figure 4. Threshold voltage for domain formation and dielectric alignment for MBBA–EBCA mixtures as a function of the dielectric anisotropy $\Delta\varepsilon$, $H = 5{\cdot}7$ kG, $T = 32$°C and sample thickness = 40 μm.

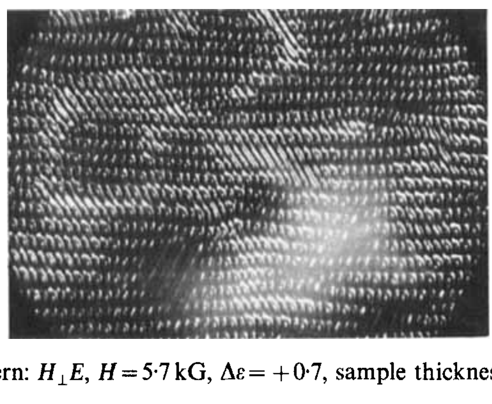

Figure 5. Grid pattern: $H_{\perp}E$, $H = 5{\cdot}7$ kG, $\Delta\varepsilon = +0{\cdot}7$, sample thickness = 250 μm, $T = 32$°C.

concluded that this is a conductive instability that exists between the values of $\Delta\varepsilon$ from $+0{\cdot}47$ to $+1{\cdot}28$ for moderately thick samples. This conductivity instability is suppressed in thin samples, in agreement with earlier investigation [1, 18].

4. Conclusions

It is evident from the experimental investigations that a stabilizing magnetic field will extend the existence of EHD instability in positive $\Delta\varepsilon$ nematics in agreement with theoretical conclusions reported earlier [16]. The theoretical and experimental values of the threshold voltages for domain formation and dielectric alignment show near agreement. The differences between the two may be due to the lack of data at different concentrations of the MBBA–EBCA nematic mixtures for the theoretical calculations. Hence the observations made are of a more qualitative importance. The new type of EHD instability in the form of a grid pattern and observed only for moderately thick samples is a conductive instability.

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