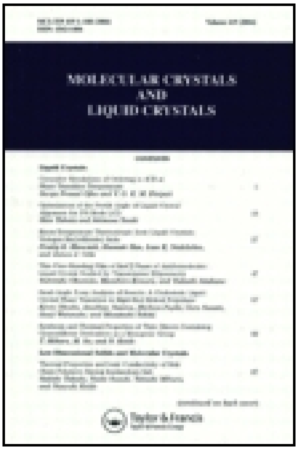

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Electrohydrodynamic Instability in Nematic Liquid Crystal Mixtures with Positive Dielectric Anisotropy

P. R. Kishore [a]

[a] Physics Department, S. V. H. College of Engineering, MACHILIPATNAM, 521 001, INDIA

Version of record first published: 20 Apr 2011.

Mol. Cryst. Liq. Cryst., 1985, Vol. 128, pp. 75–87
0026-8941/85/1282-0075/$20.00/0

Electrohydrodynamic Instability in Nematic Liquid Crystal Mixtures with Positive Dielectric Anisotropy

P. R. KISHORE

Physics Department, S. V. H. College of Engineering, MACHILIPATNAM 521 001, INDIA

(*Received August 13, 1984*)

Electrohydrodynamic instability in nematic liquid crystal mixtures of MBBA and EBCA with positive dielectric anisotropy has been investigated. Domain formation and their stability have been related to the magnitude of positive dielectric anisotropy. The sequence of various instabilities with respect to the variation of positive dielectric anisotropy of the nematic mixtures has been reported. The variation of threshold voltage for domain formation and dielectric alignment with dielectric anisotropy has been studied at moderately thick and thin samples. The instabilities observed in moderately thick and thin samples have been compared.

INTRODUCTION

Electrohydrodynamic (EHD) instabilities in nematic liquid crystals with positive dielectric anisotropy have already been the subject of much interest, both theoretical and experimental. A brief review of the previous results regarding the existence of various instabilities like Williams domains,[1] dynamic scattering[2] and dielectric alignment[3] in positive $\Delta\epsilon$ nematics has been given here.

Gruler and Meier[4–5] reported domain formation in less positive $\Delta\epsilon$ nematics upto a cut-off frequency f_c in the homogeneous geometry. Above f_c, reorientation of the optical axis was reported which is a general phenomenon for positive $\Delta\epsilon$ nematics. This EHD instability in positive $\Delta\epsilon$ nematics was explained in accordance with Carr-Helfrich theory.[6] Theoretically Gruler[7] investigated the threshold behaviour of Williams domains and dielectric alignment with dc excitation in homeotropic and homogeneously oriented samples with different signs and magnitudes of the dielectric anisotropy. Theoretical predictions of Helfrich regarding domain formation and dielectric alignment were verified. A theoretical value of $\Delta\epsilon \simeq +0.4$ was obtained, above which EHD instability does not exist. This agrees with most of the experimental values reported.

Flint and Carr[8] reported domains and dynamic scattering in positive $\Delta\epsilon$ nematics with ac and dc fields and this has been attributed to the effect of conductivity anisotropy. Recently Carr et al[9] related domain formation and their stability in nematic mixtures with positive dielectric anisotropy to the relative effectiveness of the torques associated with the conductivity and dielectric anisotropies. This investigation involved very thick samples and the results suggest the existence of EHD instability upto a value of the dielectric anisotropy $\Delta\epsilon \simeq +1.0$. P. R. Kishore et al[10] reported similar type of observations as mentioned above in MBBA-MBCA mixtures with moderately thick samples.

Domain formation and light scattering were also observed[11-12] in high positive $\Delta\epsilon$ materials like EBCA ($\Delta\epsilon \approx +20$) with dc fields. W. H. de Jeu et al[13-14] reported the existence of EHD instability in positive $\Delta\epsilon$ nematics. The same author[15-16] also investigated different types of instabilities in the same sample with different signs and magnitudes of the dielectric anisotropy. For less positive $\Delta\epsilon$ region, domains are observed at a threshold voltage and disappear at higher voltages. For this region, de Jeu suggested for a two dimensional model. In this work, domains are predicted for a value of $\epsilon_{\parallel}|\epsilon_{\perp} < 1.05$.

Penz[17] reported a two dimensional treatment for the boundary value problem to get additional information for the less positive $\Delta\epsilon$ region with dc excitation. For calculation based on the Helfrich model, MBBA values were used and $\Delta\epsilon$ was changed by keeping the other material constants constant. The calculated threshold which involves dielectric and conductivity effects agrees with the Freedericksz expression $V_c = (K_{\parallel}/\epsilon_o\Delta\epsilon)^{1/2}$ above a dielectric constant ratio $\epsilon_{\parallel} | \epsilon_{\perp} = 1.2$.

EHD instability in positive $\Delta\epsilon$ materials was also reported by Barnik et al.[18] The dependence of the threshold voltage on $\Delta\epsilon$ under constancy of other parameters was investigated. The experimental data is in agreement with the two dimensional theory of S. A. Pikin[19-20] in which boundary conditions are taken into account for Helfrich model. EHD instability had been observed about a value of $\Delta\epsilon \simeq +0.4$ above that only dielectric alignment occurs.

Zenginoglou et al[21] investigated numerically in two dimensions, the ability of homogeneously aligned nematics obtained by a magnetic field with positive $\Delta\epsilon$ to exhibit Williams domains. The results suggest that domain formation can be extended in positive $\Delta\epsilon$ nematics by the application of a stabilizing magnetic field.

Theoretically Dubois-Violette et al[22] predicted an instability with planar configuration in the positive $\Delta\epsilon$ nematics. Smith et al[23] investigated theoretically, the dynamics of EHD instability in nematic liquid crystals with planar orientation on the basis of one dimensional time dependent model of Dubois-Violette et al. From the theory, it follows that conduction regime can be suppressed altogether by using very thin samples, then only dielectric instability is possible. This had been shown for weakly positive $\Delta\epsilon$ nematics.

Alan Sussman[24] reported EHD instability in nematic and weakly cholesteric liquid crystals of positive dielectric anisotropy. He pointed out that it is possible to shift from conductivity instability to dielectric instability by reducing the sample thickness, if the sample conductance is constant. Several authors[25–27] reported the existence of EHD instability in positive $\Delta\epsilon$ nematics.

From the above theoretical and experimental investigations, the type of instability in positive $\Delta\epsilon$ nematics primarily depends on the relative magnitudes of conductivity and dielectric anisotropies. It also depends on the experimental conditions like sample thickness, type of alignment (homogeneous or homeotropic), magnetic field if used, and type of electric field (ac or dc). Most of the previous work regarding EHD instability in positive $\Delta\epsilon$ nematics was confined to thin samples except Carr's[9] work on very thick samples. The previous results suggest a value of $\Delta\epsilon \simeq +0.5$ in thin samples, below which EHD instability is likely to exist.

The present work is aimed at investigating the pcssible instabilities in positive $\Delta\epsilon$ nematic mixtures of MBBA (p-methoxy benzylidene p′-n-butyl aniline) and EBCA (p-ethoxy benzylidene p′-Cyano aniline) in the presence of electric and magnetic fields with moderately thick (250 μm) samples. For comparison, EHD instability studies have also been carried out in thin (40 μm) and oriented samples.

EXPERIMENTAL

Dielectric constant measurements are made at a frequency of the order of 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 crystal controlled oscillator as mentioned in our previous publication.[10] We believe that 100 KHz frequency is well below the dispersion region of MBBA-EBCA mixtures since the concentration of EBCA at which the dielectric anisotropy of the mixture changes sign nearly agrees with the value reported earlier[27] at 1 KHz on MBBA-EBCA mixtures.

The MBBA and EBCA samples have been prepared in our laboratory using standard procedures. MBBA has been subjected to fractional distillation under reduced pressure and EBCA has been recrystallized twice before use. The concentration of EBCA in MBBA used in this work is very low, so the viscosity of the mixture would not be much different from that of MBBA. ac conductivities of the mixtures are of the order of 10^{-10} Ohm^{-1} Cm^{-1}. The conductivity ratio $\sigma_{\parallel} \mid \sigma_{\perp}$ is 1.3 for MBBA, measured at 80Hz. This shows slight increase as the EBCA concentration increases.

A uniform and moderate sample thickness of 250 μm is used for the present work. Sample thickness of 40 μm is employed for the experimental investigation in thin samples. Homogeneous alignment in thin samples is obtained by rubbing technique. The threshold voltages in thin samples are measured by using photo multiplier and the optical observations are made with a microscope.

RESULTS AND DISCUSSION

(A) Dielectric constants of MBBA-EBCA mixtures

The dielectric constants $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ have been measured for nine samples of varying concentration of EBCA in MBBA at 100 KHz and 32°C, sample thickness = 250 μm. A magnetic field of 5.7 KG is used to align the molecular axes parallel and perpendicular to the electric field.

Figure 1 shows a plot of dielectric anisotropy $\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp}$ of the mixtures as a function of concentration of EBCA in MBBA by weight percentage at 100 KHz. The points are experimental data and the solid line is calculated assuming additivity relation.[28] The variation of $\Delta\epsilon$ of MBBA-EBCA mixtures, has been reported earlier.[27-29] The dielectric anisotropies of MBBA and EBCA are -0.41 at 32°C and $+20.0$ at 100 KHz respectively. The resulting values of $\Delta\epsilon$ calculated from the additivity relation differ with the experimental data, this may be a temperature effect.[28] The point of dielectric isotropy occurs nearly at 1.9% by weight of EBCA in MBBA.

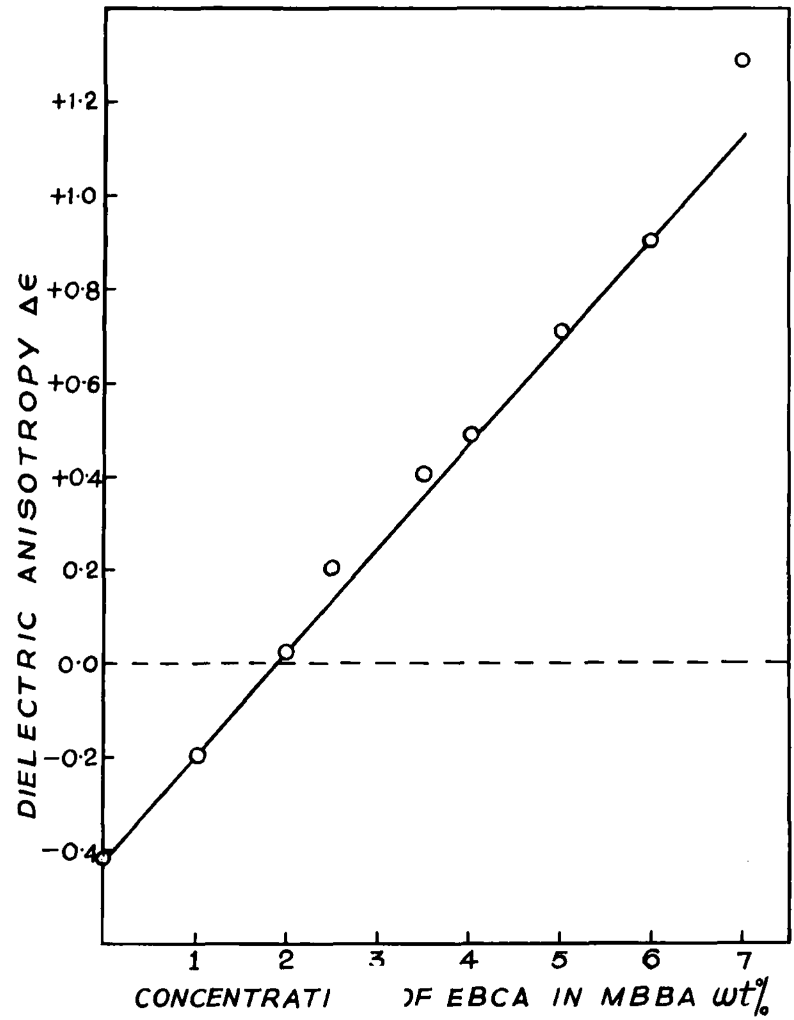

FIGURE 1 Dielectric anisotropy, $\Delta\epsilon$ of MBBA-EBCA mixtures as a function of weight percentage of EBCA in MBBA at 100 KHz. Temperature = 32°C and H = 5.7 KG.

(B) Optical observations

The instabilities in nematic liquid crystal mixtures of MBBA-EBCA with positive dielectric anisotropy at different magnitudes of $\Delta\epsilon$ in the H $\perp$ E condition is given here. Sample thickness = 250 μm.

Stable domains and strong dynamic scattering are observed for 2% concentration of EBCA in MBBA with $\Delta\epsilon$ = +0.02 at 100 KHz and

32°C. The above optical observations at +0.02 can be considered as optical effects at $\Delta\epsilon \approx O$ region.

Figure 2 shows stable domains for 2.5% concentration of EBCA in MBBA by weight with $\Delta\epsilon = +0.2$ at 100 KHz and 32°C. Stable domains are followed by weak dynamic scattering.

Figure 3 shows unstable domains for 4% concentration of EBCA

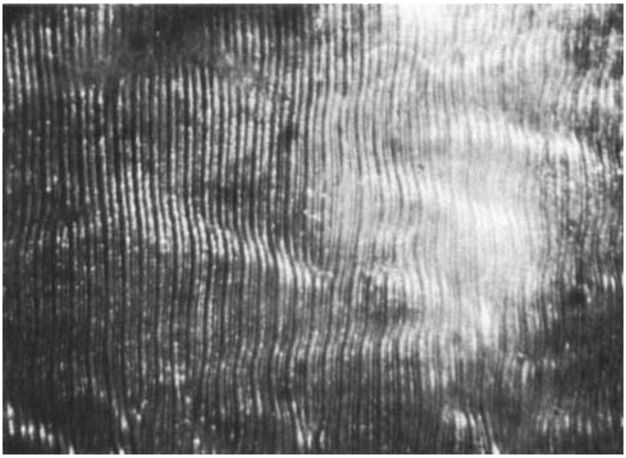

FIGURE 2 Stable domains. H $\perp$ E, H = 5.7 KG, $\Delta\epsilon = +0.2$ at 100 KHz, Temperature = 32°C.

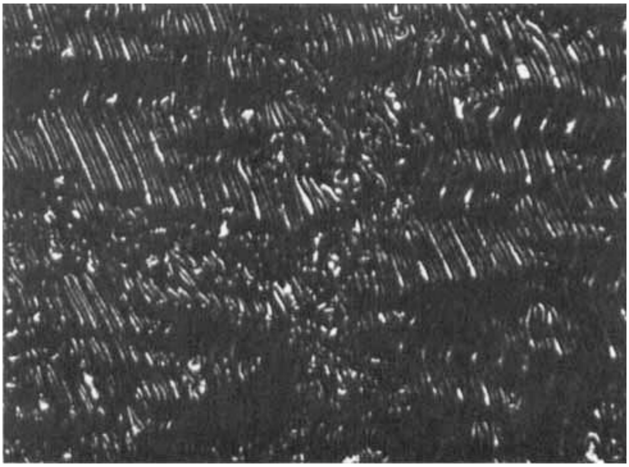

FIGURE 3 Unstable domains. H $\perp$ E, H = 5.7 KG, $\Delta\epsilon = +0.47$ at 100 KHz, Temperature = 32°C.

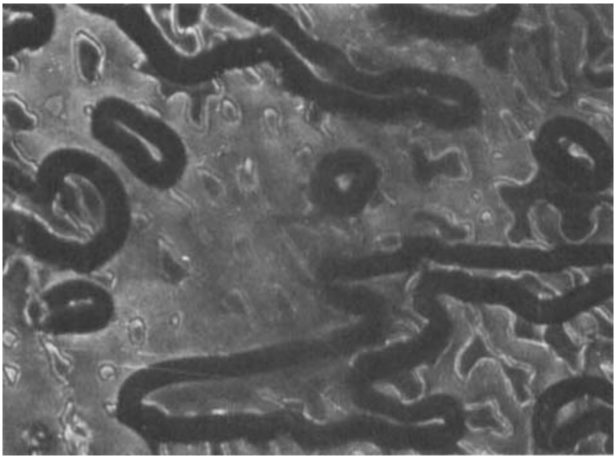

FIGURE 4 Loop domains. H ⊥ E, H = 5.7 KG. $\Delta\epsilon$ = +0.47 at 100 KHz, Temperature = 32°C.

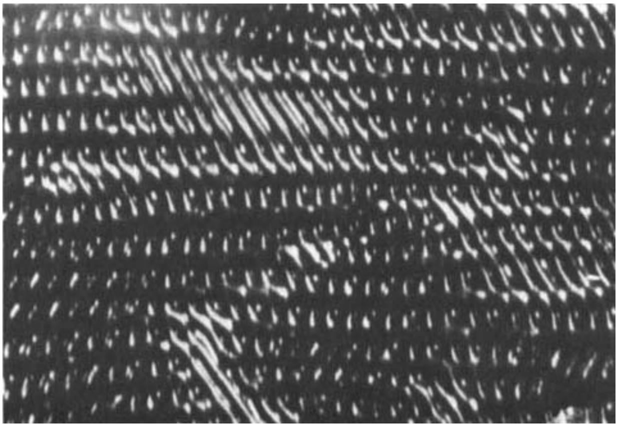

FIGURE 5 Grid Pattern. H ⊥ E, H = 5.7 KG. $\Delta\epsilon$ = +0.7 at 100 KHz, Temperature = 32°C.

in MBBA by weight with $\Delta\epsilon$ = +0.47 at 100 KHz and 32°C. As the voltage increases, these unstable domains disappear leaving the effect of reorientation. In the process of reorientation, loop domains[14] are observed as a transient effect as shown in Figure 4.

Figure 5 shows grid pattern for 5% concentration of EBCA in MBBA by weight with $\Delta\epsilon$ = +0.7 at 100 KHz and 32°C. The grid pattern is followed by reorientation. For 7% concentration of EBCA in MBBA by weight with $\Delta\epsilon = +1.28$ at 100 KHz and 32°C, only pure reorientation has been observed.

Recently Carr et al[9] related the domain formation and their stability in positive $\Delta\epsilon$ nematics to the relative effectiveness of the conduction and dielectric torques. The author of this paper reported[10] the existence of EHD instability in positive $\Delta\epsilon$ nematics relating with the molecular alignment changes in MBBA-MBCA mixtures. The same type of study has been done on MBBA-EBCA mixtures also and the results agree with that of Carr et al.[9] The molecular alignment studies on MBBA-EBCA mixtures have not been included here in view of the repetition.

(C) Variation of threshold voltage with $\Delta\epsilon$

Figure 6 represents the variation of the threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) with dielectric anisotropy measured at 100 KHz for MBBA-EBCA mixtures, sample thickness = 250 μm. The line 1 represents variation of V_{th} at 40 Hz with dielectric anisotropy in the H ⊥ E condition. The curve 2 illustrates the variation of V_F at 3 KHz with positive $\Delta\epsilon$ in the H ⊥ E condition.

It is evident from Figure 6 that V_F decreases with the increasing values of positive dielectric anisotropy. Both V_{th} decreases gradually as $\Delta\epsilon$ changes from -0.41 to $+1.28$. As the positive values of $\Delta\epsilon$ increases, the threshold difference between V_{th} and V_F decreases and finally reaches a near value at $+1.28$. According to the optical observations, the positive $\Delta\epsilon$ values are divided as follows:

(a) $\Delta\epsilon = +0.02$ stable domains and strong DSM.
(b) $\Delta\epsilon < +0.4$ stable domains and weak DSM.
(c) $+0.4 < \Delta\epsilon < +0.47$ unstable domains and reorientation.
(d) $+0.47 < \Delta\epsilon < +1.28$ Grid pattern and reorientation.
(e) $\Delta\epsilon \geqslant +1.28$ pure reorientation.

The above data indicates that EHD instability exists upto a value of $\Delta\epsilon = +1.28$, and domain formation below $+0.47$. This agrees nearly with the values reported[10] earlier on MBBA-MBCA mixtures with moderately thick samples. The variation of threshold voltage with dielectric anisotropy has been investigated by Gruler[7] and Barnik et al,[18] which agrees with the present work. The dielectric constant ratio $\epsilon_{\parallel} \mid \epsilon_{\perp} = 1.2$ at 100 KHz for 7% concentration agrees with the theoretical value of Penz.[17]

For comparision, EHD instability studies are also extended to thin homogenously oriented samples (40 μm) with the same MBBA-EBCA

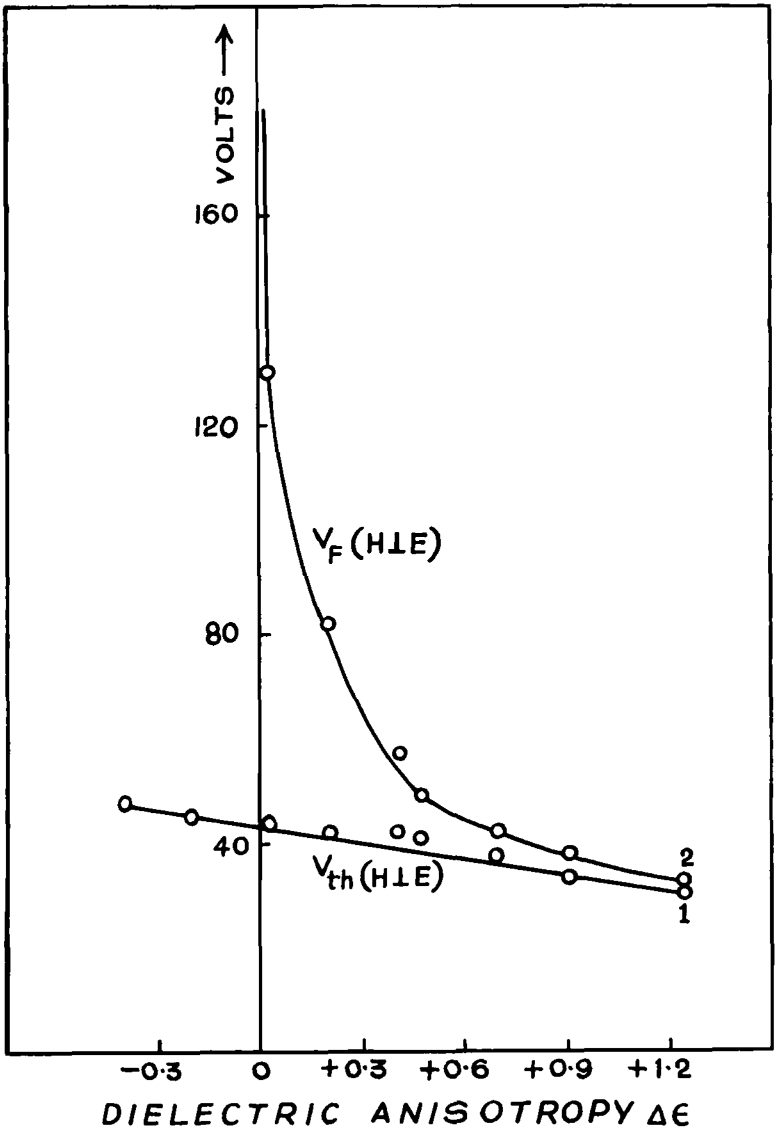

FIGURE 6 The threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) in MBBA-EBCA mixtures as a function of the dielectric anisotropy $\Delta\epsilon$ at 100 KHz. H = 5.7 KG, Temperature = 32°C and sample thickness = 250 μm.

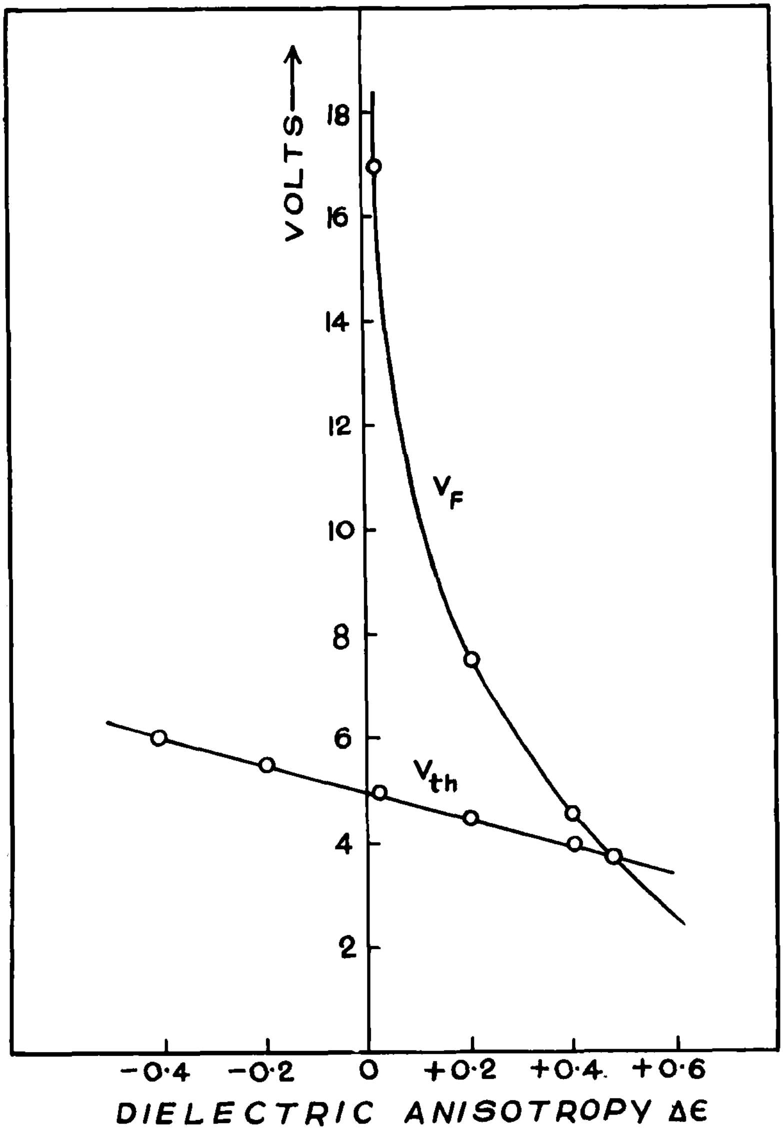

FIGURE 7 The threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) in MBBA-EBCA mixtures as a function of the dielectric anisotropy $\Delta\epsilon$ at 100 KHz. H = 0, Temperature = 32°C and sample thickness = 40 μm.

mixtures. Figure 7 shows the variation of V_{th} at 40 Hz and V_F at 3 KHz with $\Delta\epsilon$. At the low values of $\Delta\epsilon \simeq +0.1$, stable domains and dynamic scattering are observed. Between +0.1 and +0.47, unstable domains and reorientation exist. At and above the values of +0.47, there is only reorientation.

The above results indicate that domain formation in moderately thick and thin samples exist upto a value of positive $\Delta\epsilon = +0.47$. Domain formation and EHD instability exist upto the same value of positive $\Delta\epsilon$ in thin samples. But in the case of moderately thick samples, the EHD instability extends from +0.47 to +1.28 in the form of grid pattern a shown in Figure 5.

It is evident from the above results that the conductivity instability which plays vital role to cause EHD instability has been suppressed in thin samples to the lower values of positive $\Delta\epsilon$ as compared with moderately thick samples. Recent theoretical work of Smith et al[23] and experimental observations of Alan Sussman[24] also suggest that the conductivity instability can be suppressed in thin samples.

Figure 8 shows the variation of V_{th} at 40 Hz and V_F at 3 KHz with dielectric anisotropy in H $\perp$ E condition, sample thickness = 40 μm. The values of V_{th} and V_F show an increase with the magnetic field. The maximum value of $\Delta\epsilon$ below which domain formation and EHD instability exists is +0.52, which is greater than the value obtained in the absence of magnetic field (Figure 7). The existence of EHD instability to higher values of positive $\Delta\epsilon$ is due to the effect of stabilizing magnetic field according to Zenginoglou et al.[21]

Acknowledgments

The author is grateful to Prof. C. R. K. Murty and Prof. E. F. Carr for their kind interest and encouragement.

References

1. R. J. Williams, *J. Chem. Phys.*, **39**, 384 (1963).
2. G. H. Heilmeier, L. A. Zanoni and L. A. Barton, *Proc. Inst. Elect. Electronics Engrs.*, **56**, 1161 (1968).
3. V. Freedericksz and V. Zolina, *Trans. Faraday Soc.*, **29**, 919 (1933).
4. H. Gruler and G. Meier, *Mol. Cryst. Liq. Cryst.*, **12**, 289 (1971).
5. H. Gruler and G. Meier, *Mol. Cryst. Liq. Cryst.*, **16**, 299 (1972).
6. W. Helfrich, *J. Chem. Phys.*, **51**, 4092 (1969).
7. H. Gruler, *Mol. Cryst. Liq. Cryst.*, **27**, 31 (1974).
8. W. T. Flint and E. F. Carr, *Mol. Cryst. Liq. Cryst.*, **22**, 1 (1973).
9. E. F. Carr, W. T. Flint and J. H. Parker, *Phys. Rev.*, A, **11**, 1732 (1975).

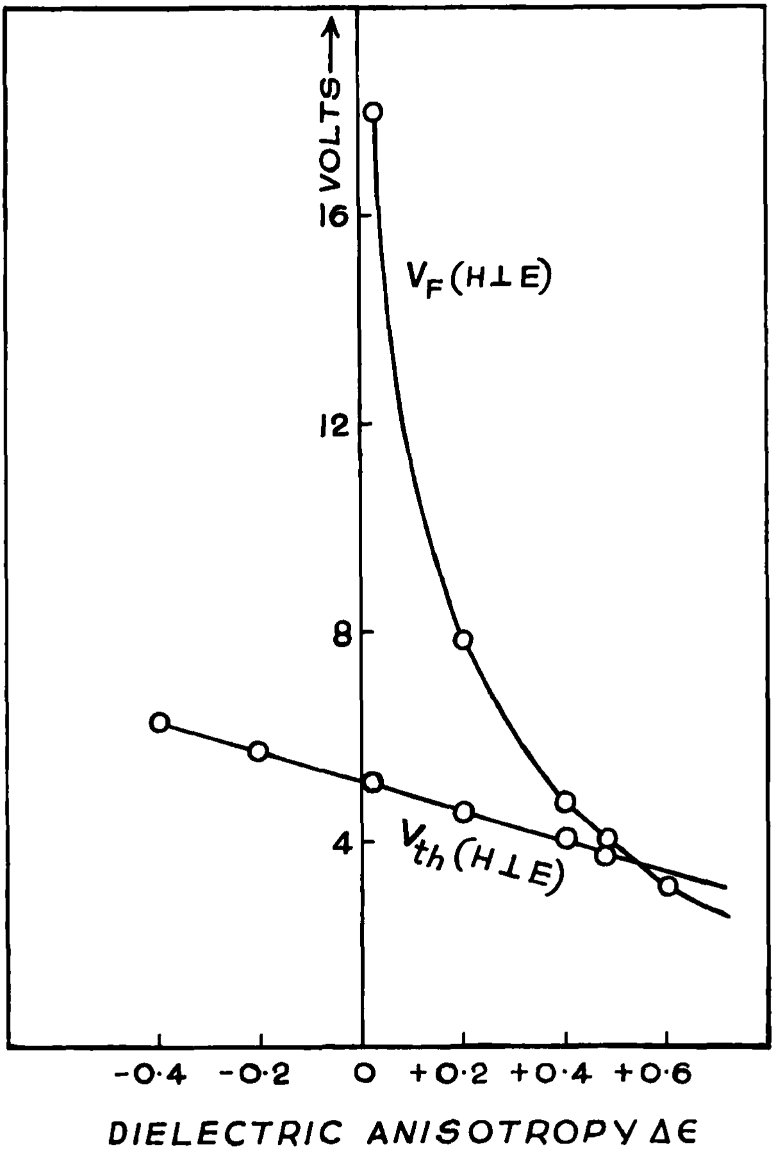

FIGURE 8 The threshold voltage for domain formation (V_{th}) and dielectric alignment (V_F) in MBBA-EBCA mixtures as a function of the dielectric anisotropy $\Delta\epsilon$ at 100 KHz. H = 5.7 KG. Temperature = 32°C, and sample thickness = 40 μm.

10. P. R. Kishore and C. R. K. Murthy, *Mol. Cryst. Liq. Cryst.*, (in Press).
11. W. H. de Jeu, C. J. Gerritsma and A. M. Van Boxtel, *Phys. Lett.*, **34A**, 203 (1971).
12. A. I. Baise and M. M. Labes, *J. Chem. Phys.*, **59**, 551 (1973).
13. W. H. de Jeu and C. J. Gerritsma, *J. Chem. Phys.*, **56**, 4752 (1972).
14. W. H. de Jeu, C. J. Gerritsma and Th. W. Lathouwers, *Chem. Phys. Lett.*, **14**, 5031 (1972).
15. W. H. de Jeu, C. J. Gerritsma, P. Van Zanten and W. J. A. Goossens, *Phys. Lett.*, **39A**, 355 (1972).
16. W. H. de Jeu and Th. W. Lathouwers, *Mol. Cryst. Liq. Cryst.*, **26**, 235 (1974).
17. P. A. Penz, *Mol. Cryst. Liq. Cryst.*, **23**, 1 (1973).
18. M. I. Barnik, L. N. Blinov, M. F. Grebenkin, S. A. Pikin and V. G. Chigrinov, *Sov. Phys. JETP*, **42**, 550 (1975); *Phys. Lett*, **51A**, 175 (1975).
19. S. A. Pikin, *Sov. Phys. JETP*, **33**, 641 (1971).
20. S. A. Pikin and A. A. Shtol' berg, *Sov. Phys. Crystallogr*, **18**, 283 (1973).
21. H. M. Zenginoglou and I. A. Kosmopoulos, *Mol. Cryst. Liq. Cryst.*, **43**, 265 (1977).
22. E. Dubois-Violette, P. G. de Gennes and O. Parodi, *J. Phys.*, **32**, 305 (1971).
23. I. W. Smith, Y. Galerne, S. T. Lager Wall, E. Dubois-Violette and G. Durand, *J. de Physique*, **36**, C1 - 237 (1975).
24. A. Sussman, *Appl. Phys. Lett.*, **29**, 633 (1976).
25. A. Takase, S. Sakgami and M. Nakamizo, *Jap. J. Appl. Phys.*, **14**, 228 (1974).
26. B. Kerllenevich and A. Coche, *Electron, Lett.*, **11**, 17 (1975).
27. R. A. Kashnow and H. S. Cole, *Mol. Cryst. Liq. Cryst.*, **23**, 329 (1973).
28. R. E. Michel and G. W. Smith, *J. Appl. Phys.*, **45**, 3234 (1974).
29. D. Meyerhofer, *J. Appl. Phys.*, **46**, 5084 (1975).