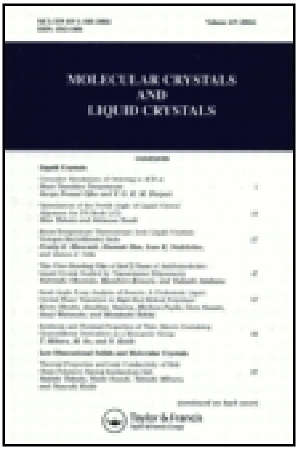

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Comparative Optical Studies of Electrohydrodynamic Instabilities in Selected Nematic Liquid Crystals with some Smectic Ordering

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A broken Williams Domain or square texture has been observed when low frequency a.c. electric and d.c. magnetic fields are applied to OOCP (di-1′,4″-octyloxybenzylidene-1,4-diaminochlorobenzene). This texture has been observed by Berman and Gelerinter in Eastman dynamic scattering mixture II. X-ray studies by de Vries indicate the presence of some smectic ordering in some alkoxybenzylidene-1,4-diaminochlorobenzenes, especially OOCP. The conditions required for the occurrence of the square texture are presented in detail and compared with those obtained by Berman and Gelerinter in KII. Experimental variables include sample temperature and thickness, magnetic and electric field intensities and frequency of the applied electric field. Agreement of the observations with those predicted for Williams Domains is presented and compared with similar observations on compounds having nematic and smectic phases. Agreement of the texture periodicity variation versus magnetic field with that predicted by Pikin and Shtol'berg is also reported.

INTRODUCTION

Berman, Gelerinter and deVries[1] have recently reported observing a broken Williams Domain in Eastman dynamic scattering mixture II (KII). Previously Rondelez[2] has reported a series of conductivity measurements and texture observations. When examining di-4′,4″-decyloxybenzylidene-1,4-diaminochlorobenzene (DOCP), he finds textures similar to the broken Williams Domains observed both in this work and in Ref. 1. DOCP has a negative dielectric anisotropy, a positive conductivity anisotropy[2] and

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smectic order fluctuations in the nematic.[3] Studies[2] by Rondelez of materials with negative conductivity anisotropies indicate textures different from those observed in KII and di-4′,4″-octyloxybenzylidene-1,4-diaminochlorobenzene (OOCP). OOCP has a negative dielectric anisotropy, and smectic order fluctuations throughout its nematic phase.[3] The amount of smectic ordering decreases with increasing temperature. The effects of this smectic ordering also are observed in epr studies.[4,5] Because of the similarity of OOCP and DOCP we feel that OOCP probably also has a positive conductivity anisotropy. Certainly this must be true in the central and upper portions of its nematic range, and it is probably true throughout the nematic range.

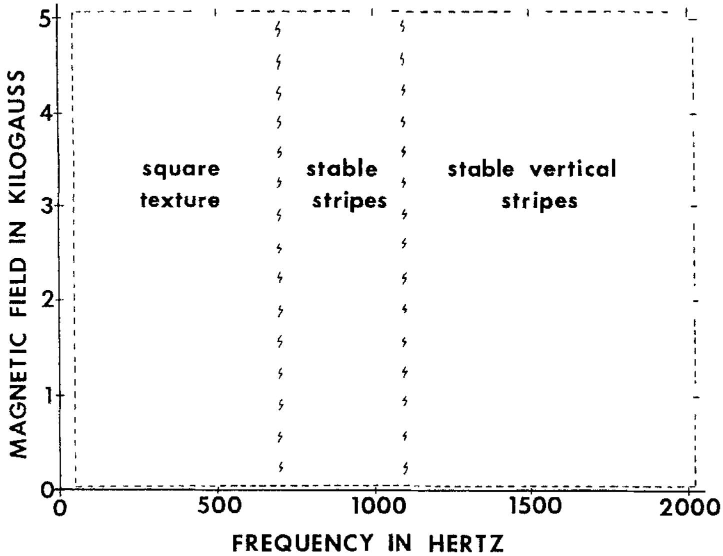
In this work we wish to report the results of a detailed study of the square texture in OOCP. Where appropriate the results will be compared to the KII results reported earlier.

EXPERIMENTAL RESULTS

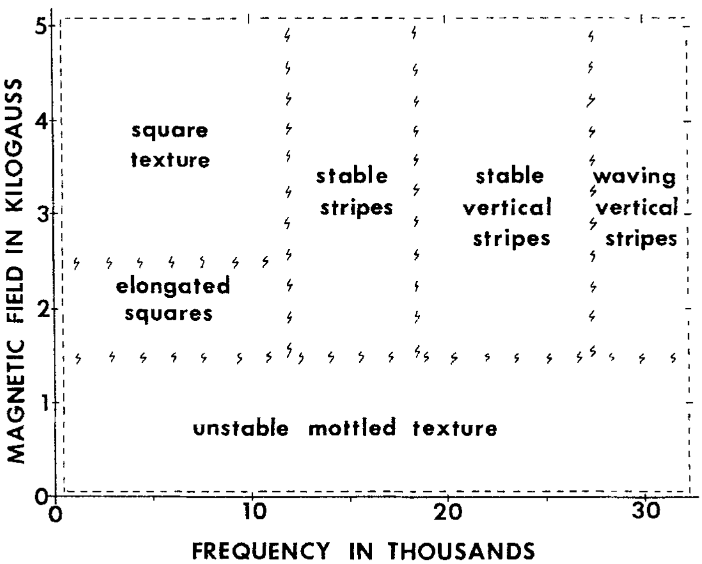
The experimental set-up is the same as reported in Ref. 1. The OOCP was synthesized at the Kent State Liquid Crystal Institute. In the form received it was quite pure and did not display any textures or dynamic scattering when subjected to a.c. electric fields. When the sample is doped with 0.33% by weight tetrabutylamoniumnitrate and 0.30% by weight *n*-hexadecylamine one can observe both the textures described and dynamic scattering. This doping is held constant for all of the experiments.

In Figure 1 we illustrate the conditions under which one observes the square and other textures. One should note that the highest frequency for which the square texture occurs increases with increasing temperature. This is consistent with the increase in crossover frequency (as shown in Figure 9) with temperature. Of much more interest is the observation of the minimum d.c. magnetic field (H) required for the square texture. In KII at room temperature one requires[1] 4 KG, in OOCP at 140°C one requires $2\frac{1}{2}$ KG, and in OOCP at 85°C one observes the texture in the residual field of the magnet ($\sim$70 G). This indicates that as the smectic order fluctuations increase, the d.c. magnetic field required for observation of the square texture decreases. The square texture aligns parallel and perpendicular to the direction of the magnetic field. Since the conducting glass plates are not rubbed, the applied magnetic field also serves to align the director when the a.c. electric field is off.

In Figure 2 we observe the threshold voltage V_{th1} versus frequency, at a sample temperature of 100°C. The curves are nearly horizontal until 3000 Hz. At this point the curves begin to rise and the square texture begins to distort



(a)



(b)

FIGURE 1 Nature of the optical textures in doped OOCP as a function of the d.c. magnetic field and the frequency of the electric field (sample thickness 150 μ).
a) Temperature = 85°C.
b) Temperature = 140°C.

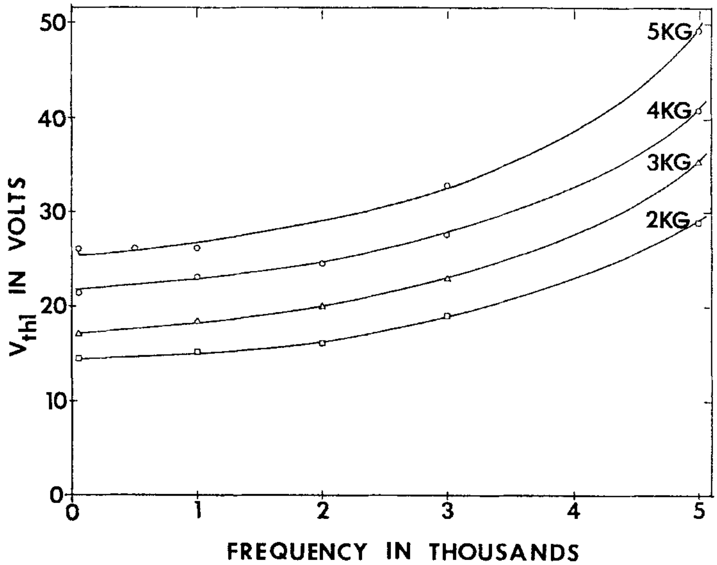


FIGURE 2 Threshold voltage for the onset of the square texture in OOCP versus the frequency of the a.c. electric field for various values of the d.c. magnetic field. $T = 100^\circ$C, $d = 164\,\mu$.

(the absolute value of the frequencies depends upon the doping). Eventually vertical stripes form at a higher frequency. A further increase in frequency results in waving vertical stripes as previously described for KII. The threshold voltage increases with increasing H. This effect is discussed in more detail later in the paper.

In Figure 3 we plot the periodicity versus frequency and observe the periodicity to be constant (within experimental error) at constant H. One also finds that the periodicity decreases with increasing field. This effect is discussed in more detail later in this manuscript.

In Figure 4 we plot sample thickness versus $V_{\text{th}1}$. Thick samples tend towards a field threshold while thin samples tend towards a voltage threshold. This has been observed[1] for KII and calculated by Dubois-Violette *et al.*[6] for Williams Domains. Their results are

$$V\text{th}(\omega, H) = V(\omega, o)\sqrt{1 + \left(\frac{H}{Ho}\right)^2},$$

where

$$Ho = \frac{\pi}{d}\sqrt{\frac{K}{\Delta\chi}}.$$

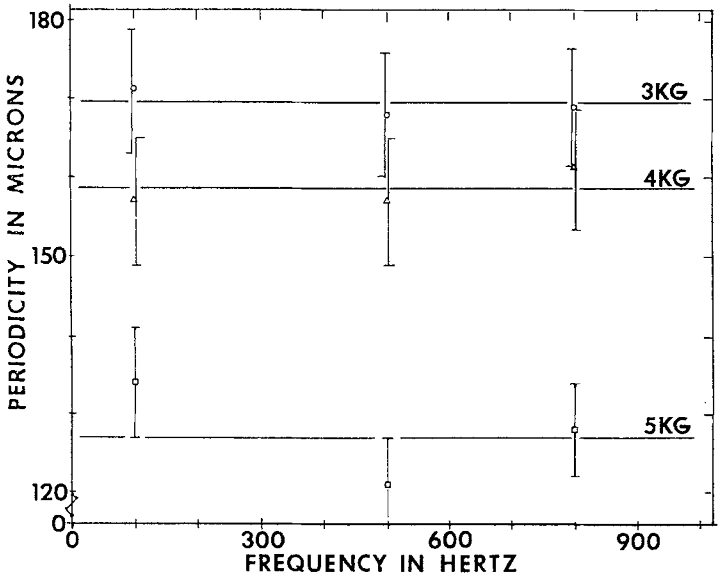


FIGURE 3 Square periodicity in OOCP versus the frequency of the a.c. electric field for various values of the d.c. magnetic field. $T = 100°C$, $d = 197\ \mu$.

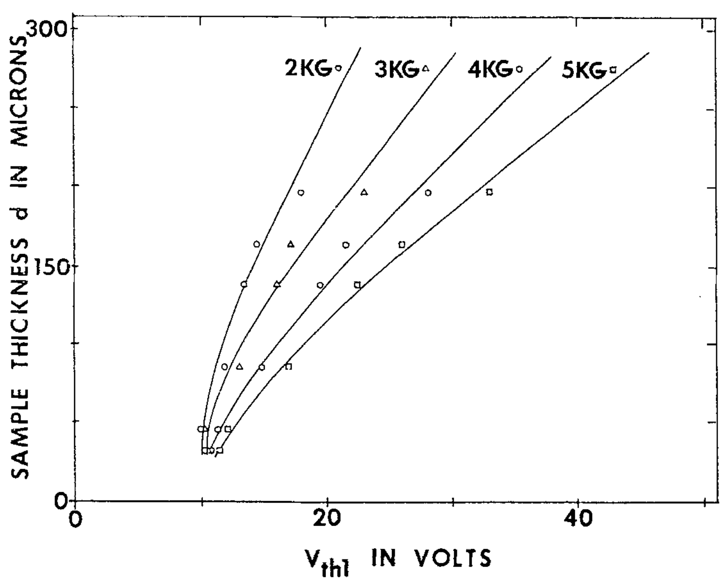


FIGURE 4 Threshold voltage for the onset of the square texture in OOCP versus sample thickness at various values of the d.c. magnetic field. $T = 100°C, f = 100$ Hz.

Combining and squaring one gets

$$V_{th}^2(\omega, H) = V^2(\omega, o) + \frac{V^2(\omega, o)H^2}{\pi^2 \dfrac{K}{\Delta\chi}} d^2.$$

So we predict that a plot of V_{th}^2 versus the sample thickness squared would result in a straight line if H is held constant (increasing H would increase the slope of the line). The slope of the line should give $K/\Delta\chi$ where K is the effective elastic constant and $\Delta\chi$ is the anisotropy in the magnetic susceptibility. The intercept gives the threshold voltage when $H = 0$. In Figures 5 and 6 we plot V_{th}^2 versus d^2 for OOCP and KII respectively. The lines are a least squares fit of a straight line. We get a fairly good fit for OOCP and a fair fit for KII. The derived parameters are listed in Table I. Note the excellent agreement between the 3-5 KG values for OOCP. There is much

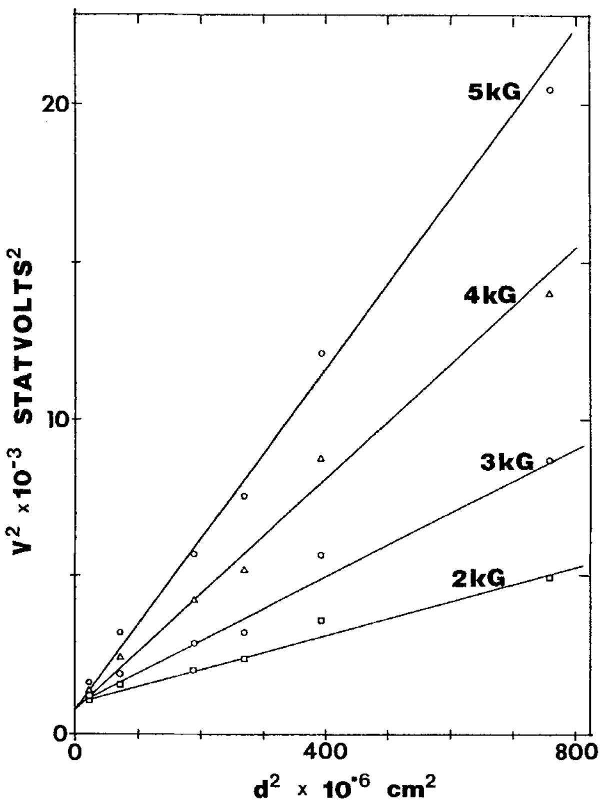


FIGURE 5 The square of the threshold voltage versus the square of the sample thickness in OOCP. (Data same as Figure 4.)

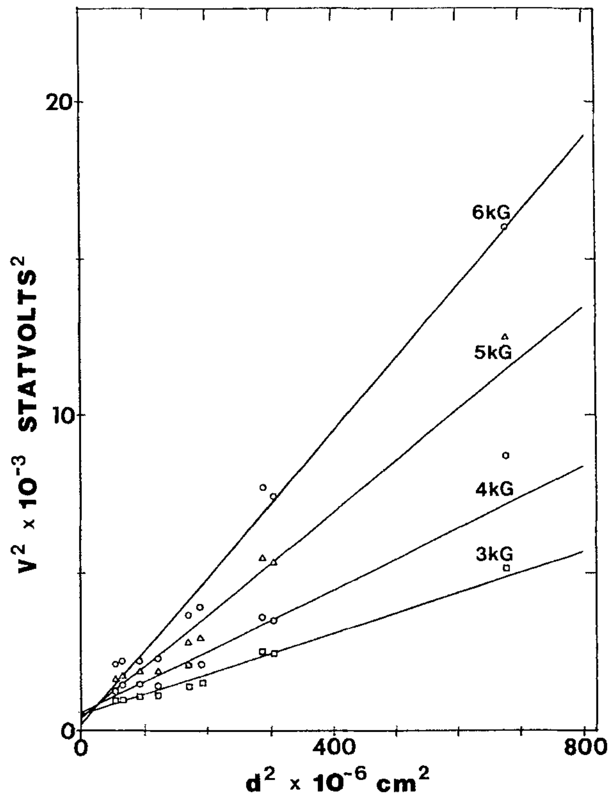


FIGURE 6 The square of the threshold voltage versus the square of the sample thickness for KII. (Data taken from Ref. 1.)

TABLE I

Experimentally measured parameters for OOCP and KII. Here $V_{H=0}$ is the threshold voltage (volts) with $H = 0$ and $\sqrt{(K/\Delta\chi)}$ is the root of the elastic constant divided by the anisotropy in the conductivity (cgs).

	OOCP		KII	
H (Gauss)	$V_{H=0}$ volts	$\sqrt{(K/\Delta\chi)}$ cgs	$V_{H=0}$ volts	$\sqrt{(K/\Delta\chi)}$ cgs
6000	—	—	4.4	5.8
5000	8.5	8.6	5.6	7.4
4000	8.5	8.2	7.0	9.6
3000	8.6	8.1	6.2	7.8
2000	9.1	7.6	—	—

more scatter in the data for KII. This is consistent with the poorer fits for KII. Also note the rather high values of $\sqrt{(K/\Delta\chi)}$. Using a different method[7] Berman and Gelerinter measured $\sqrt{(K_{22}/\Delta\chi)}$ as 1.5 for *p*-azoxyanisole in good agreement with other groups.[8–10] We believe that the higher values observed for KII and OOCP are probably due to a stiffening of the nematic due to the presence of short range smectic ordering. Another group[11] has also calculated that the threshold voltage is proportional to H in the limit of large H.

The square periodicity increases with increasing sample thickness in a manner similar to Williams Domains. In Figures 7a and b we show photographs of the textures for approximate sample thicknesses of 45 and 165 μ respectively. The texture dependence on the sample thickness is clearly illustrated. In Figure 7c we plot the periodicity versus sample thickness. Here again we observe smaller periodicity at higher H. It is interesting to note that, at 1 KG, the periodicity is larger than the sample thickness over a large portion of the thickness range. A similar effect was noted[1] for a 70 μ

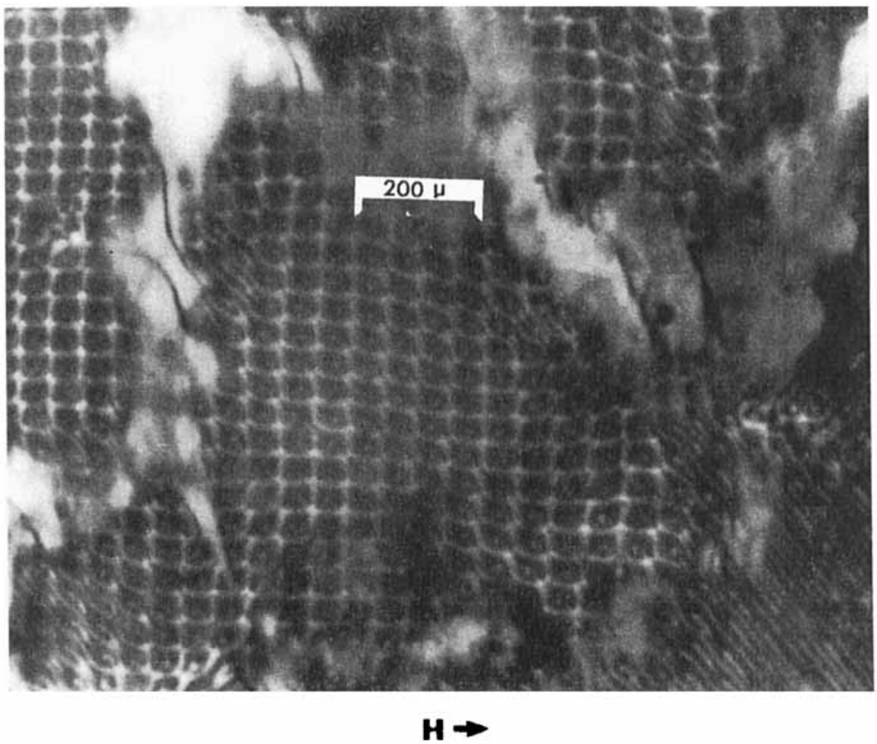


FIGURE 7a. Photograph of texture with $d \cong 45\ \mu$.

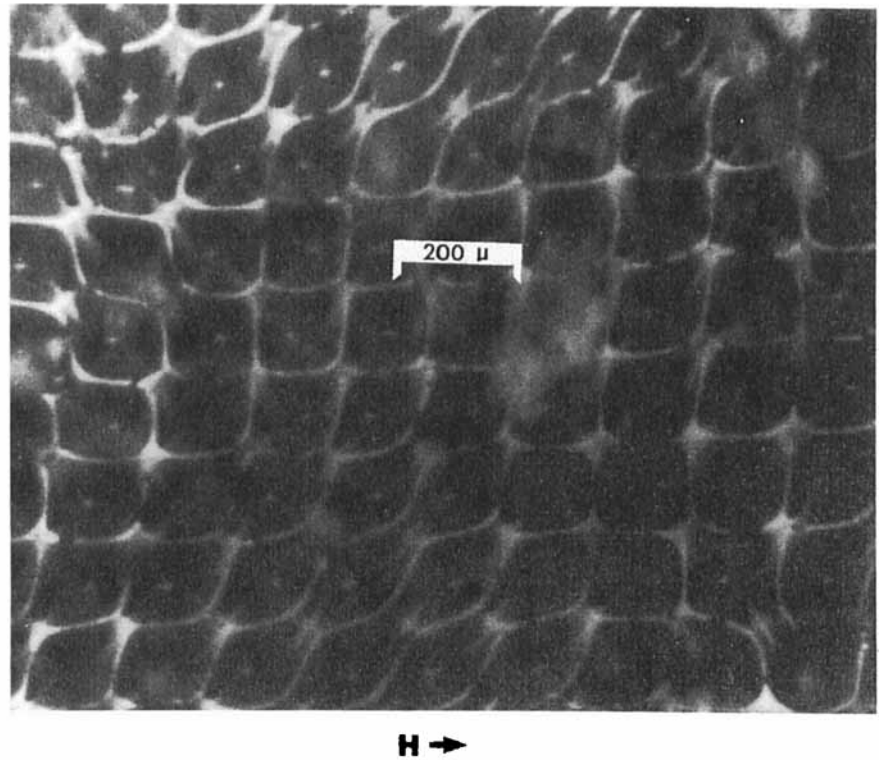


FIGURE 7b Photograph of texture with $d \cong 165\ \mu$.

sample of KII at 3 KG. Since we see the effect for both liquid crystals we are no longer able to blame it on experimental error as was done in Ref. 1.

Pikin and Shtol'berg[11] have written a theory of electrohydrodynamic effects in liquid crystals. They predict, for large magnetic fields, that the periodicity will go as $1/\sqrt{H}$, and for low H, the periodicity will vary slowly (decreasing with increasing H). In Figure 7d we plot the periodicity versus $1/\sqrt{H}$ for different sample thicknesses. The points corresponding to 3, 4 and 5 kG are on straight lines going through the origin. At lower fields the variation is much less as predicted.[11]

As the OOCP sample temperature is raised one finds the square periodicity (Figure 8) decreasing linearly. This is different from what we observed[1] for KII. In the latter case the periodicity remained essentially constant from 25 to 60°C and then started to decrease.

In Figure 9 we plot the threshold voltage versus frequency for OOCP at different temperatures. We note that the critical frequencies increase sharply

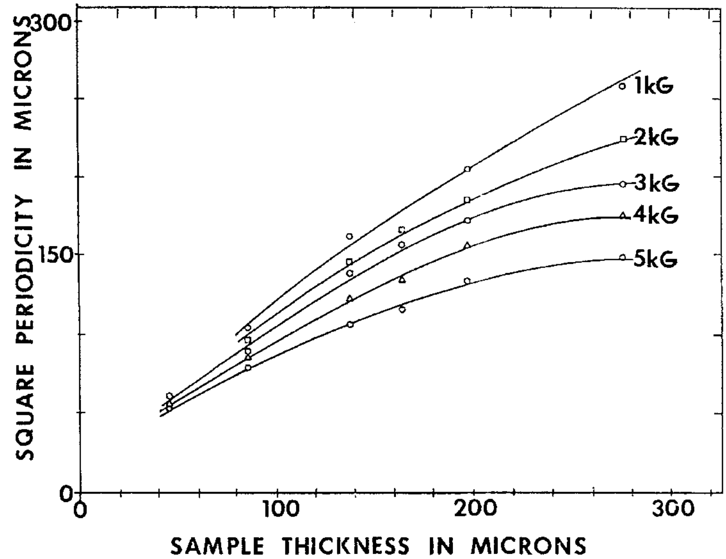


FIGURE 7c Square periodicity versus sample thickness (OOCP, T = 100°C, f = 100 Hz).

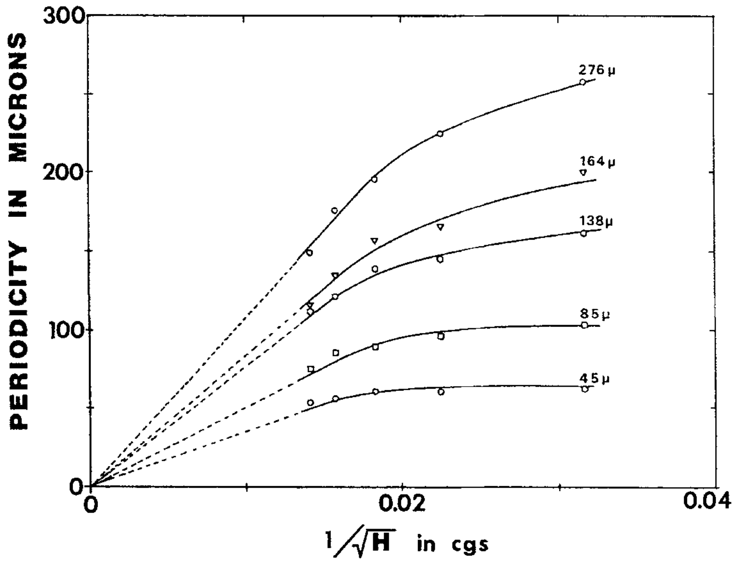


FIGURE 7d Square periodicity in OOCP versus the reciprocal of the square-root of the d.c. magnetic field for various sample thicknesses.

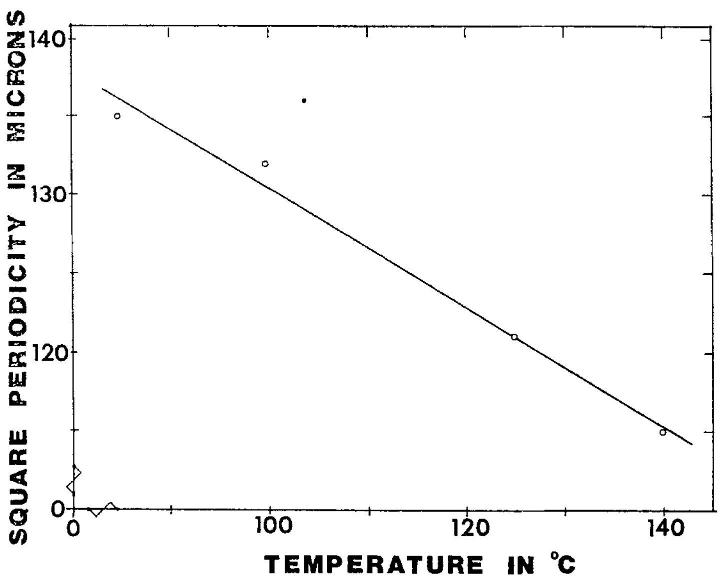


FIGURE 8 Domain width versus temperature. (OOCP, H = 5 KG, d = 164 μ).

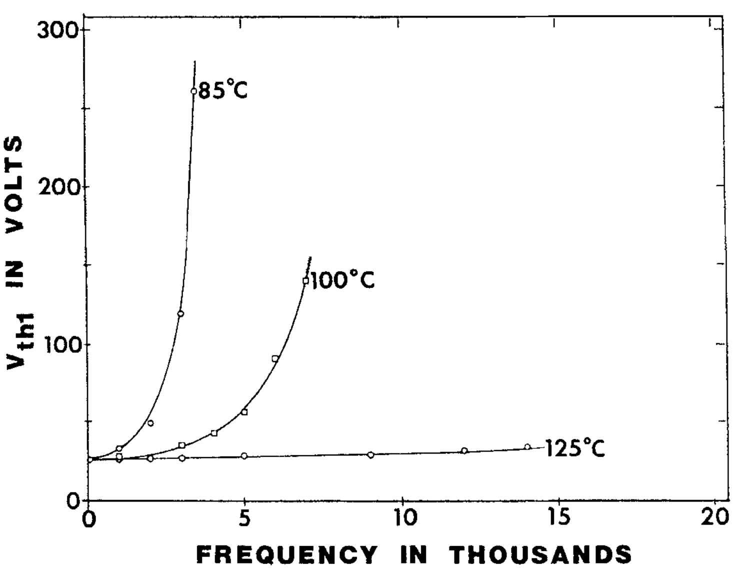


FIGURE 9 Threshold voltage versus frequency for different temperatures.

with temperature as previously noted for KII. The large values for the critical frequency are due to the sample doping.

DISCUSSION

Three conditions seem to be necessary for observing the square texture in a nematic material. They are a negative dielectric anisotropy, a positive conductivity anisotropy, and short range smectic order. Our studies of OOCP correlate the amount of smectic order with the ease of obtaining the texture. If X is the direction of the magnetic field (and hence the undisturbed director) and Z is perpendicular to the conducting glass plates (i.e., the direction of the light beam), then the texture becomes very dim or disappears when the incoming light is polarized parallel to the Y direction. This leads us to believe that the director does not tilt out of the XZ plane. Our picture of the distortion is a bend wave in the XZ plane whose amplitude varies with Y. This gives a twist axis along the Y direction. The distortion looks like an egg crate so that the valleys of the distortion focus the light into stripes along the X and Y directions and the peaks of the high amplitude bend wave focuses the light into a dot in the middle of the squares. It would be of interest to calculate the free energy of this bend and twist distortion and compare it to a pure bend distortion. Such a calculation would require a knowledge of the bend and twist elastic constants, however.

The strong similarities between our observations and those predicted for Williams Domains leads us to believe that we are observing an electrohydrodynamic instability. It is interesting to note the consistent measurements of $\sqrt{(K/\Delta\chi)}$ for OOCP. The large variation in the KII results are somewhat disappointing. The large values are a reflection of the smectic ordering present.[12]

We have not tried to observe the square texture in a nematic with short range smectic A ordering. We plan to look for a nematic above a smectic A phase that satisfiies the three conditions listed in the beginning of this discussion and to try to observe the texture in that material.

Acknowledgements

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