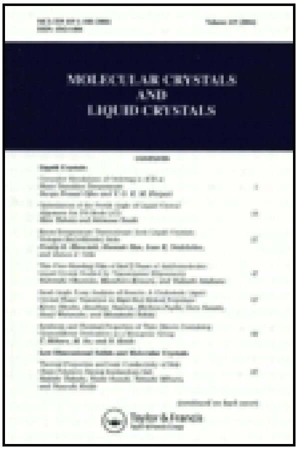

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Optical Studies of Electric Field Effects in Nematic Liquid Crystals That Have Some Smectic Ordering

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Optical Studies of Electric Field Effects in Nematic Liquid Crystals That Have 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 Eastman Dynamic Scattering Mixture II or di-4′,4″-octyloxybenzylidene-1,4-diaminochlorobenzene. In the absence of the magnetic field ordinary Williams Domains are observed. The square pattern is only observed in the lower temperature portion of the nematic range. Evidence is presented for the existence of smectic ordering in this region. The conditions required for the occurrence of the square pattern are presented in detail. The features of the pattern are studied versus magnetic field, temperature and frequency. Many of our observations are in approximate agreement with those predicted for Williams Domains. Some are not. Our observations are compared with similar observations on compounds having nematic and smectic phases. It is clear that a three dimensional theory is required to completely understand the observations.

INTRODUCTION

In the last several years there has been a large amount of study,[1–5] both experimental and theoretical, of nematic liquid crystals undergoing hydrodynamic instabilities. Most of the experimental studies used the room temperature nematic N-(*p*-methoxybenzylidene)*p*′-*n*-butylaniline (MBBA) which displays normal Williams Domains when subjected to low frequency electric fields. There have also been studies of materials which have both a nematic and smectic phase.[6,7] In these cases the authors report observing a broken Williams Domain or square pattern when the sample, which is at a temperature just above the nematic–smectic transition, is subjected to a low frequency electric field. Presumably the presence of pretransitional smectic ordering is responsible for the transverse flow causing a combination bend, twist and splay wave to be stabilized instead of the bend wave one usually associates with normal Williams Domains.

We wish to report observing a similar texture under somewhat different conditions. We are able to reproducibly observe the texture in the nematic di-4′,4″-octyloxybenzylidene-1,4-diaminochlorobenzene (BOCP) and the nematic mixture Eastman Dynamic Scattering Mixture II (KII). The bulk of our experiments were performed on KII because of its convenient room temperature nematic phase. However, BOCP displayed the same general results as those observed for KII. Eastman Chemicals[8] supplied values for the dielectric constants and conductivities of KII. We report below the results and the conditions for which the square pattern appears. We will also present evidence for the existence of smectic ordering in these materials. We believe that this smectic ordering is necessary for one to reliably observe this texture.

EXPERIMENTAL RESULTS

The typical experimental set-up is shown in Figure 1. The sample is placed between conducting glass plates. Incoming white light is polarized parallel to the undisturbed director. The light is passed through the sample and the analyzer which is adjusted for maximum contrast. The applied electric field

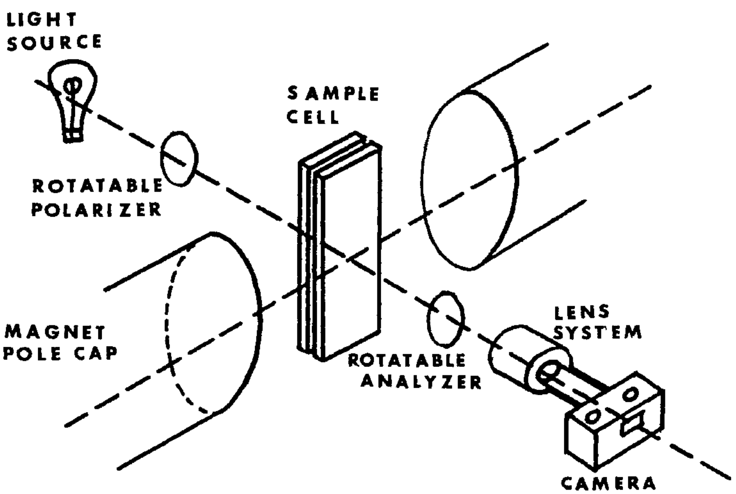

FIGURE 1 Sample geometry. The sample cell contains a homogeneously aligned nematic liquid crystal.

is parallel to the light path. In addition one can apply a magnetic field perpendicular to the electric field.

When a few volts at a frequency of 100 hertz is applied to a rubbed sample of KII at room temperature (RT) one observes normal Williams Domains. However, if a magnetic field is also applied one observes the texture illustrated in Figure 2. We will refer to the texture as the square pattern. One should note the collection of squares and the short stripes going off diagonally. The latter are at an angle of 35° to the vertical.

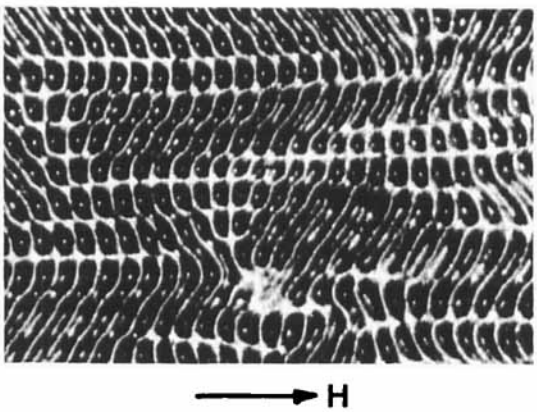

FIGURE 2 Texture observed in KII at room temperature when $f = 100$ Hz and $H = 6$ KG.

In Figure 3 we illustrate the conditions under which one observes the square pattern. One requires an applied electric field at a frequency above a few tens of hertz but less than 700 hertz. A magnetic field greater than 4 KG is also required. The pattern is observed up to 6.5 KG which is the limit of our present magnet. Above 700 hertz the transition to the high frequency textures occur. These are currently under study and will be reported on at a later time.

The square texture occurs with lines approximately parallel and perpendicular to the magnetic field. This is independent of the initial alignment of the director. For example, if a well rubbed sample is rotated about an axis parallel to the electric field, then the pattern remains stationary. An unrubbed sample and a randomly rubbed sample also produce patterns parallel to the magnetic field (B). We conclude that B, and not the surface forces, aligns the director when the square pattern is observed.

The threshold voltage (V_{th1}) at RT for the onset of the square pattern is plotted versus frequency for different magnetic fields in Figure 4. One observes that V_{th1} increases with increasing B. This is what is normally expected for Williams Domains.[1] At lower frequencies V_{th1} is approximately

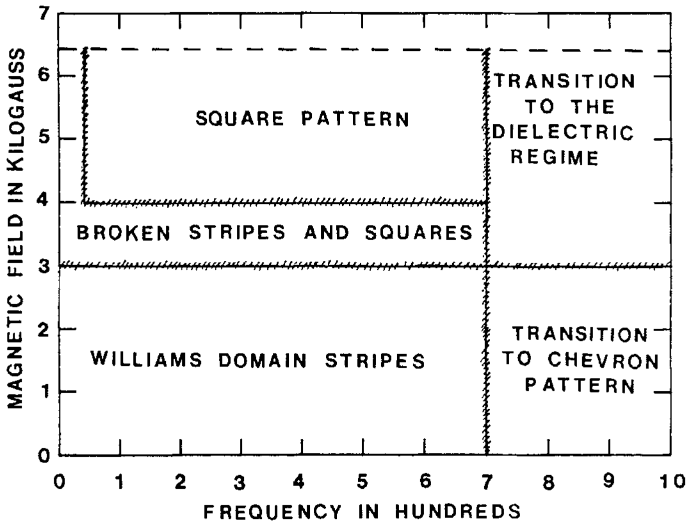

FIGURE 3 Nature of the texture in KII at room temperature as a function of the strength of the applied magnetic field and the frequency of the electric field.

constant. Above 700 hertz V_{th1} begins to rise. This rise is accompanied by a distortion of the square pattern. These effects are indicative of the start of the approach towards the critical frequency. The square pattern is observed for voltages up to approximately 2V above the threshold after which dynamic scattering gradually takes over.

In Figure 5 one sees a plot of the periodicity (vortex size) of the pattern versus frequency for different magnetic fields. Within the scatter of the points the periodicity is independent of the frequency, but the periodicity increases with decreasing B. Above approximately 700 hertz the squares elongate with the width remaining approximately constant. At high frequency (about 2000 Hertz) the texture consists of stripes. These stripes appear to undergo a waving motion. If no magnetic field is applied the chevrons, described in the literature,[3] appear.

It is also of interest to note the value of V_{th1} versus sample thickness. The question one might ask is does the instability have a voltage or field threshold. The results of such a study are illustrated in Figure 6. One should note the voltage threshold to a thickness of approximately 110 microns. At larger thicknesses the plot is a straight line indicating a field threshold. In the limit of high magnetic field, other authors[1] have predicted a field threshold for Williams Domains. The requirement is that $(B/B_0)^2 \gg 1$. B_0 can be estimated from $V_{th1}(w, B)/V_{th1}(w, 0)$ as approximately 1000 gauss so that $(B/B_0)^2$ goes from approximately 36 to 9. It is not very surprising to see a field

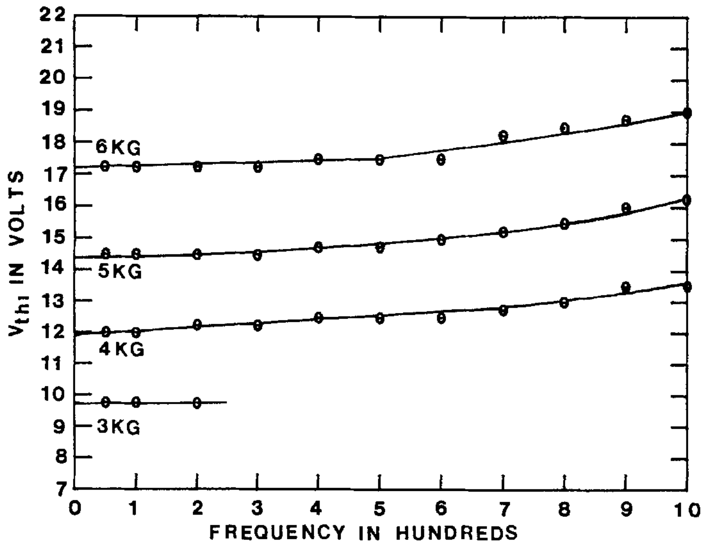

FIGURE 4 The threshold voltage for the square pattern in KII at room temperature as a function of the strength of the applied magnetic field and the frequency of the electric field.

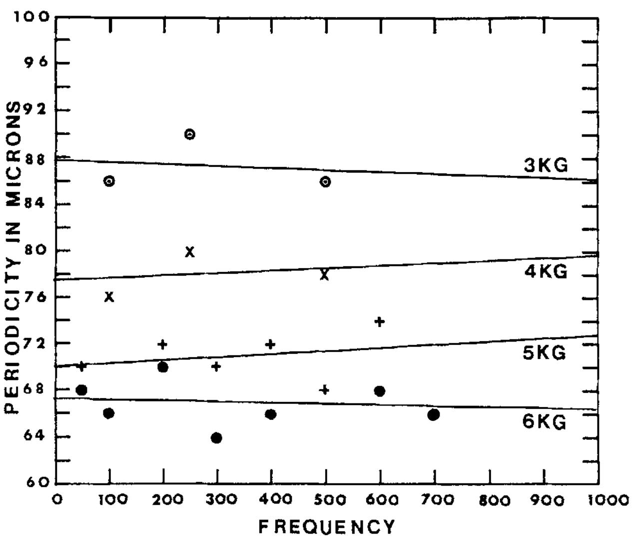

FIGURE 5 The width of the square domain in KII at room temperature as a function of the strength of the applied magnetic field and the frequency of the electric field.

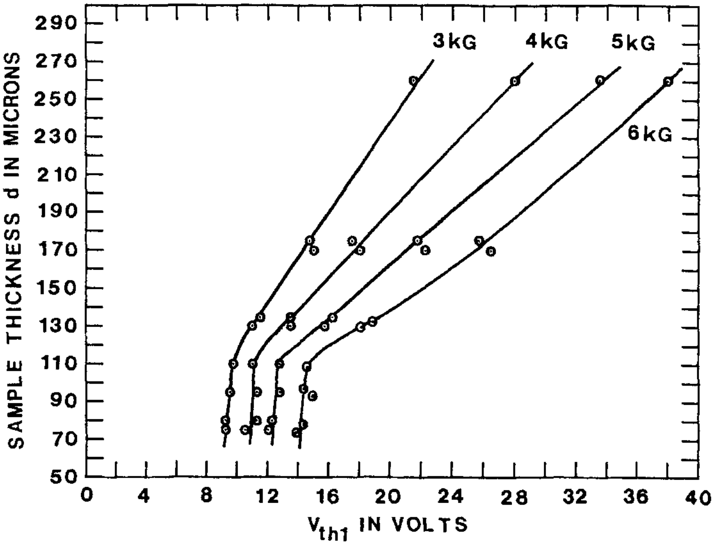

FIGURE 6 The threshold voltage for the square pattern in KII at room temperature as a function of the sample thickness and the strength of the applied magnetic field.

threshold, but we are surprised to note the crossover to a voltage threshold below $d = 110$ microns.

In Figure 7 we plot the periodicity of the square pattern (e.g. the vortex size) versus the sample thickness at RT. When normal Williams Domains ($B = 0$) occur the vortices essentially fill the sample thickness. We have previously shown (Figure 5) that the B field causes the size of the vortices to decrease. In Figure 7 we see that the size of the vortices is somewhat smaller than the sample thickness over most of the range. The square pattern periodicity at 3 KG for a 70 micron sample appears to be slightly larger than the sample thickness. This probably has to do with error in measuring the sample thickness which is about 10% in this range.

When the temperature of KII is raised one finds the square pattern elongating to waving stripes perpendicular to B (Figure 8). The stripes look like ordinary Williams Domain stripes. The waving or rocking motion of the stripes causes them to break and rejoin with nearby stripes. The width of the patterns (squares at low temperature, stripes at high temperature) were measured versus temperature and are plotted in Figure 9. There is about a 20% decrease in width over the range.

The threshold voltage versus frequency was measured for KII at three temperatures, and the results are shown in Figure 10. One notes the sharp rise in critical frequency with temperature. A similar result was observed by

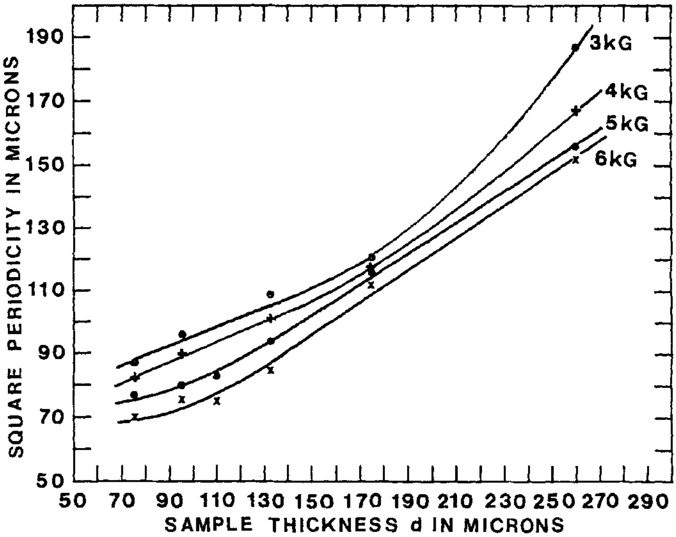

FIGURE 7 The width of the square domains in KII at room temperature as a function of the sample thickness and the strength of the applied magnetic field.

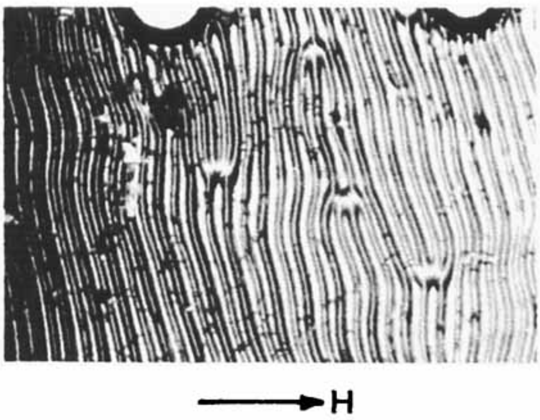

FIGURE 8 Texture observed in KII at a temperature of 87°C when $f = 100$ Hz and $H =$ 6 KG.

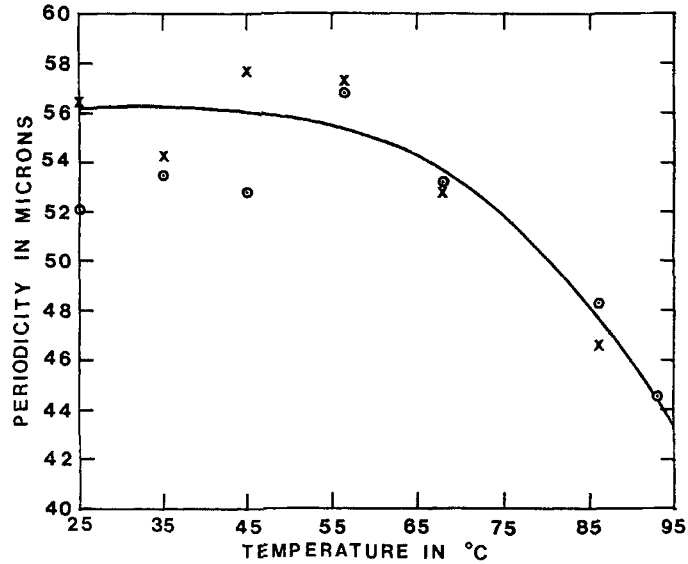

FIGURE 9 The width of the domains in KII as a function of temperature.

Rondelez[6] when he studied di-4′,4″-decyloxybenzylidene-1,4-diaminochlorobenzene (DOCP). The critical frequencies are rather high. This is indicative of the fact that impurities are present which are probably added to make the mixture more suitable for dynamic scattering. The data of Figure 10 was taken at $B = 6$ KG, but we have measured the critical frequency versus B and find that B does not change the observed critical frequency.

Finally, we have observed the textures displayed by BOCP under similar conditions but have not repeated the detailed measurements made on KII. We observe a square texture in the lower temperature region of the nematic phase followed by rocking stripes in the high temperature region. It should be mentioned that rocking stripes have been observed[9] in MBBA by other authors and are attributed to a dynamically stable intermediate state between Williams Domains and Dynamic Scattering.

DISCUSSION

Textures similar to the square pattern we are reporting are observed without the use of an applied magnetic field, by Rondelez[6] in DOCP and by Goscianski and Leger[7] (G. & L.) in N-(*p*-*n*-butoxybenzylidene)*p*′-*n*- octylaniline (40.8) and 4′-,4″-di-*n*-octyloxyazoxybenzene (C8). These com-

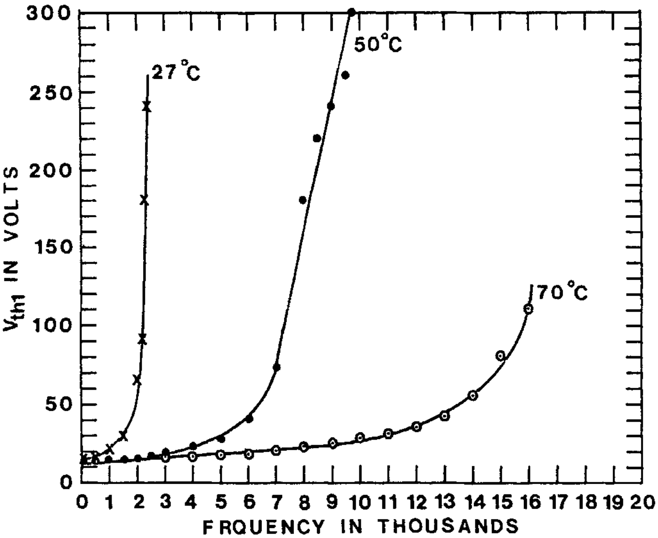

FIGURE 10 The threshold voltage for the instability in KII as a function of the frequency of the electric field and the temperature.

pounds, unlike KII and BOCP, have smectic phases as well as nematic phases. Normal Williams Domains are observed in 40.8 and C8 at high temperatures. Presumably, the hydrodynamic flow causes twist instabilities giving rise to "broken" Williams Domains. These authors observe the Helfrich parameter to be close to one just above the N–S transition and attribute this to smectic order fluctuations. Also the conductivity ratio $\sigma_{\parallel}/\sigma_{\perp}$ goes from above one to below one as the N–S transition is approached.

This suggests that one should perhaps look for smectic ordering in the materials of this study. There is evidence[10,11] for the existence of smectic ordering in BOCP in the literature. De Vries reports smectic ordering throughout the nematic range of BOCP with the amount of order decreasing with increasing temperature. We have performed x-ray studies of KII at room temperature and have found evidence for a small amount of smectic ordering. This is illustrated in Figure 11. In 11A we show a drawing of the x-ray diffraction pattern of a sample of KII (RT) which was aligned by a magnetic field. The dog bone shape center structure is indicative of smectic C ordering with a tilt angle (α) of 35°. (This is the same angle that the diagonal lines in our square texture make with the vertical!) In 11B we show the results that we obtain with MBBA, a normal nematic material. Another group[12] has occasionally observed broken Williams Domain patterns in MBBA but we have been unable to reproduce the texture. The x-ray results for BOCP are taken from the literature[10] and shown in 11C.

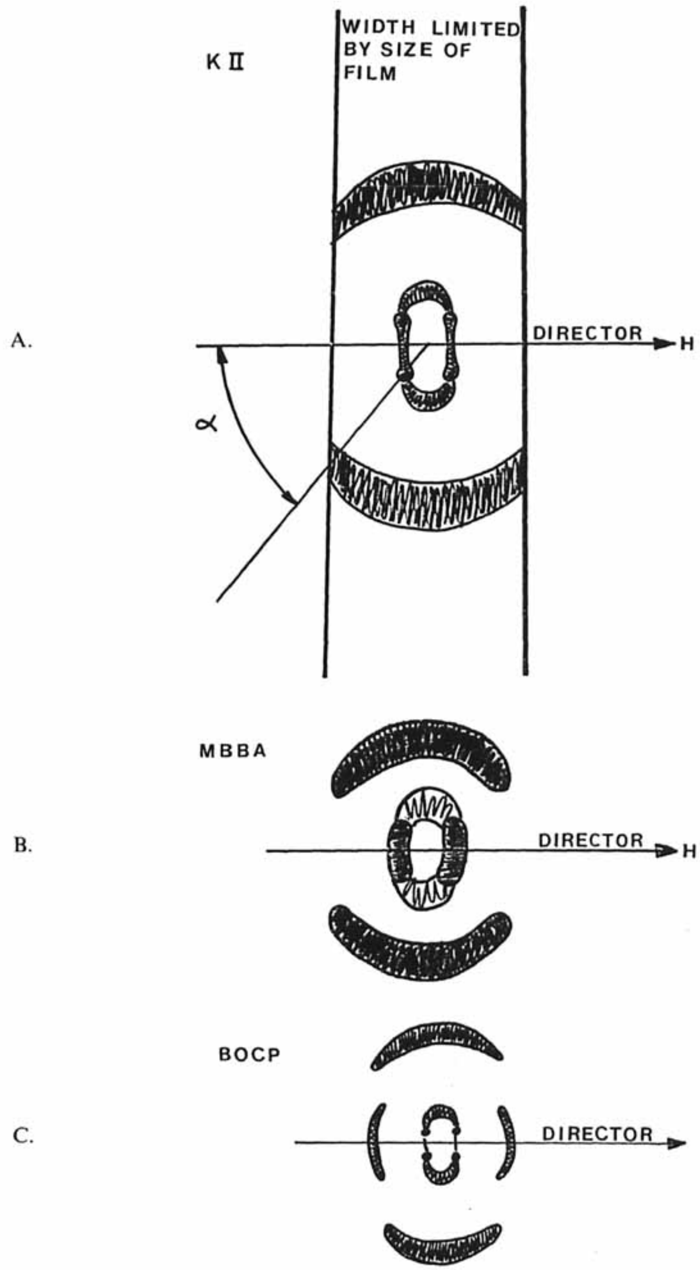

FIGURE 11 A. X-ray diffraction pattern of KII at room temperature (aligned by a magnetic field) indicates that the nematic has some smectic ordering.
B. X-ray diffraction pattern of MBBA at room temperature. Normal nematic.
C. X-ray diffraction pattern of BOCP indicating some smectic C-like ordering at the low temperature end of its nematic range.

A three-dimensional calculation is needed to completely understand our results. However, some observations are in order. The threshold voltages approximately follow what one would expect from the literature predictions.[1] The periodicity of the pattern, which is related to the wave vector of the distortion is dependent upon temperature and magnetic field. It is very slightly, if at all, dependent upon frequency. The theoretical treatment of Williams Domains does not predict this variation since it assumes a constant wave vector π/d. The distortion must be in the plane determined by the undisturbed director (N_0) and the electric field. This is evidenced by the requirement that the incoming light not be polarized perpendicular to N_0 for the texture to be seen. A strong magnetic field is necessary to observe the broken Williams Domains and smectic ordering is present in BOCP and KII. MBBA which does not have smectic ordering does not display the effect. Perhaps a mechanism similar (it clearly cannot be exactly the same) to that proposed by G. & L. is at work here, i.e. the destabilization of twist fluctuations. The magnetic field will cause the ions to move perpendicular to the molecules. (B replaces the conductivity effect of C8 and 40.8 since $\sigma_{\|} > \sigma_{\perp}$ throughout the nematic temperature range.) This, in turn, may lead to a destabilization of a twist fluctuation.

Finally, it is interesting to note that even though some smectic order exists in KII the $\sigma_{\|}/\sigma_{\perp}$ continues to increase with decreasing temperature unlike 40.8 or C8. KII is a mixture. Perhaps one of the components is approaching a smectic phase and displaying smectic order fluctuations as a pretransitional effect while other component(s) are normal nematic materials. This could explain the apparent inconsistency in the conductivity ratio. In any case, it is clear that the smectic ordering is necessary to observe the effect. As the temperature is raised, the ordering is decreased and ordinary Williams Domains are observed.

Acknowledgement

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