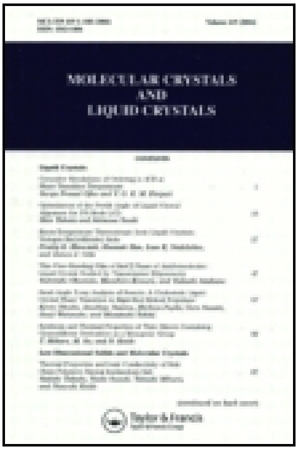

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Flow Patterns in Bulk Samples of a Nematic Liquid Crystal Due to Electric Fields†

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It is known that a mechanism exhists for producing molecular alignment in liquid crystals which is associated with the ionic conduction anisotropy and hydrodynamic flow. This flow can be observed with the help of either dyes, dust particles, or scattered light. The flow cells so observed usually extend from one electrode to the other and exhibit a domain width dependent on the strength of the applied electric and magnetic field. With increasing electric field strength, these flow cells decrease in width and are generated and dissipated more rapidly. Data relating to the average state of alignment as the result of applied electric and magnetic fields is also presented.

INTRODUCTION

A considerable amount of work has been carried out involving flow patterns in nematic liquid crystals due to electric fields. Most of this research is for sample thicknesses of approximately 100 microns or less. Most of the work reported here involves bulk samples 2 mm thick and the flow patterns were also observed in the presence of low frequency electric and static magnetic fields acting simultaneously. A model for flow patterns and molecular alignment, similar to that suggested by Helfrich[1] to explain Williams domains,[2] has been suggested[3] to explain ordering in bulk samples due to high electric and magnetic fields. This model is illustrated in Figure 1. We do not intend to imply that the alignment can be represented by a cosine function. Investigations with thin samples by other researchers[4] have provided experimental evidence for such a model.

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‡ Based on a portion of the thesis of E. J. Sinclair, to be submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Maine.

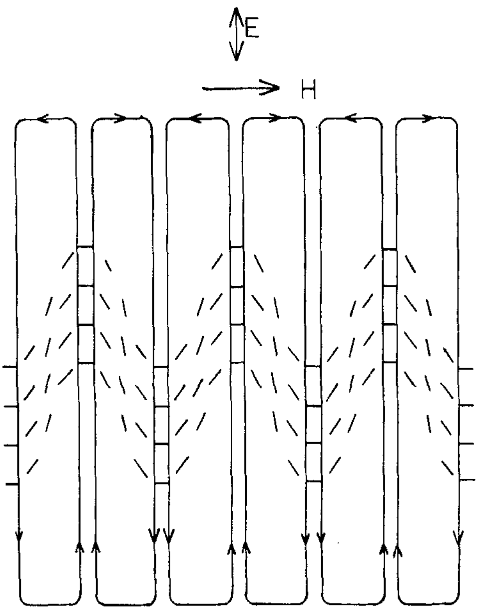


FIGURE 1 Molecular alignment in presence of magnetic and electric fields (conduction regime).

Although quantitative results are presented for the work reported here, the primary objective was to obtain qualitative information. The results vary some with the dimensions of the sample cell and they will also show some variation among different samples of *n*-(*p*-methoxybenzylidene)-*p*-butylaniline (MBBA). We have found that if the dimensions remain constant and the sample does not have time to change its properties during a run, information concerning flow patterns can be obtained. Since microwave techniques were used to obtain information concerning the average state of alignment, a cell was constructed identical to the microwave cell for the visual observations.

EXPERIMENTAL

Measurements of the microwave dielectric loss were used as a probe of the average state of alignment. The microwave techniques were similar to those reported earlier.[5] The sample cell was a section of K-band waveguide with a center plate electrically insulated from the guide. The center plate divides the cell into two sections with dimensions 2 × 8 mm for each section. The sample cell for visual observations was identical to the microwave cell except that it was open at the top for visual observations. This allowed direct observation of the surface. When a low frequency electric field of sufficient strength was applied, flow cells were created. The variation of the velocity of the fluid results in variations of the light scattered from the surface. The contrast is very poor but photographs of the surface can provide some evidence for flow cells. Photographs of the surface were obtained for different values of the electric field applied parallel and perpendicular to a magnetic field. For electric fields not far above threshold a waiting period of several minutes was required before there was some uniformity of flow cell widths.

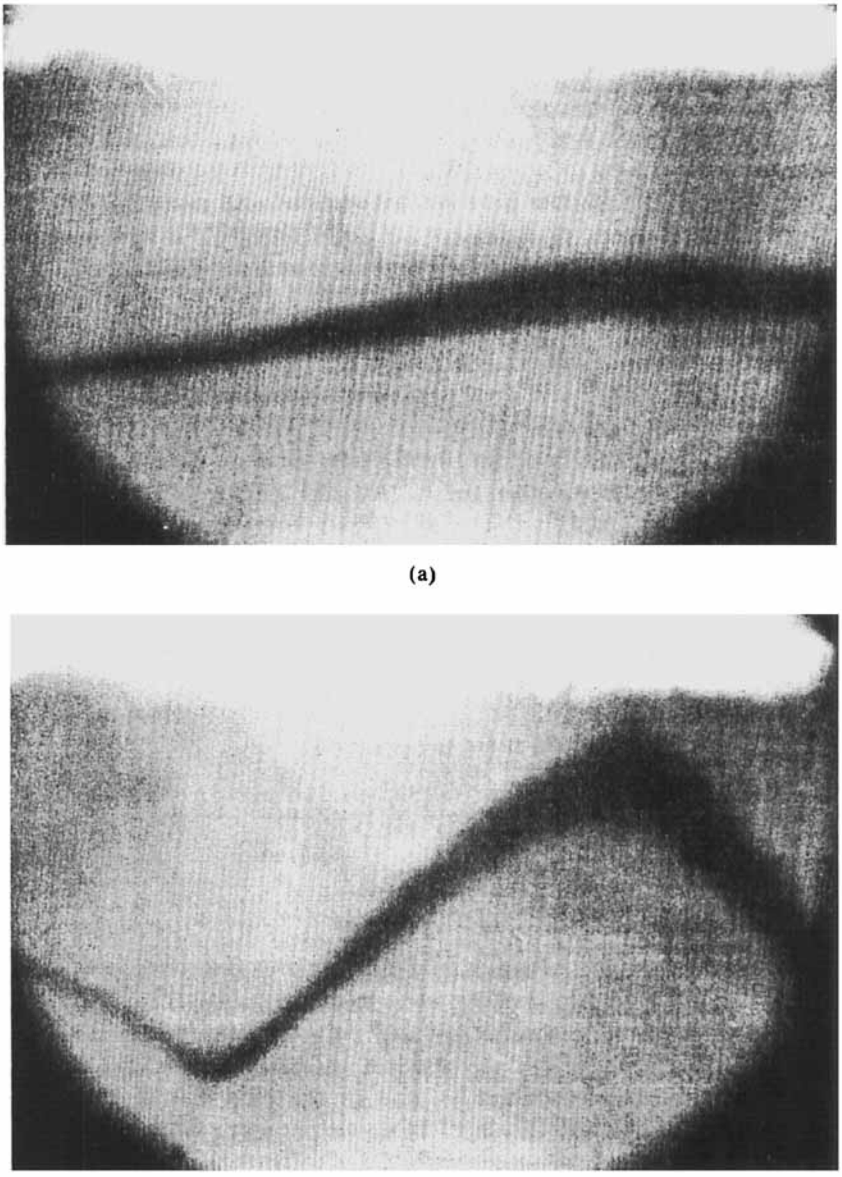
Dyes were also used to indicate flow patterns near threshold fields when the motion of the fluid was relatively slow. A dye was placed on each of the plates before applying the electric field. When the electric field was applied the material flow pulled the dye into the liquid, which created patterns that could be photographed. Near threshold fields, the dyes worked reasonably well, but at higher fields with rapid motion and narrow flow cells it mixed so quickly with the sample that photographs were not obtained. We experimented with many different dyes, but methyl violet dissolved in dimethyl formamide was used for the work reported.

MBBA was used for all the results reported here. The ratio of the electrical conductivities parallel and perpendicular to the director was 1.5. The resistivity of the material used for the visual observations was approximately 10^9 ohm-cm and that for the microwave measurements was lower. All measurements were made at room temperature.

RESULTS AND DISCUSSION

A Flow Cells Near Threshold Using Dyes

Figure 2 provides some information about possible flow cells at threshold fields. In Figure 2a, a thread of dye was placed on the surface of the sample and parallel to a 4-kG magnetic field and the conducting plates of the cell. As the applied voltage was increased, a threshold field (approximately 1000 V/cm) was reached where motion of the fluid was first observed. Shortly after reaching the threshold field, the photograph shown in Figure 2a was obtained. The bending of the thread of dye indicates a flow cell width comparable to the separation of the plates which was 2 mm. Cell widths of this size are difficult to obtain but there appears to be a tendency to form flow cell widths comparable to the plate separation at threshold fields. This is consistent with the formation of Williams domains in thinner samples. Parker[6] observed Williams domains in a cell with a 1-mm plate separation



(a)

(b)

FIGURE 2 Surface of the sample with a plate separation of 2 mm and a 4-kG magnetic field applied parallel to the conducting plates and surface of the sample.

a. Thread of dye on the surface of the sample and parallel to the magnetic field. $E = 0$.

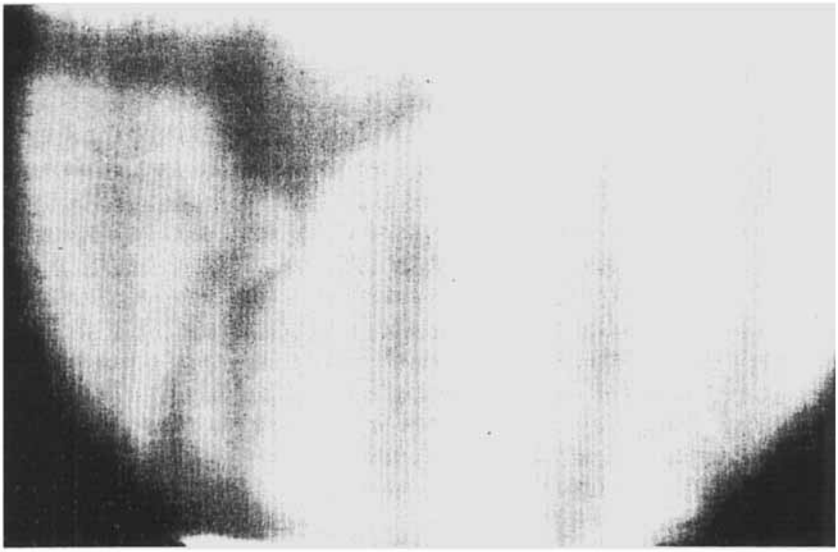
b. Distorted thread due to material flow. $E = 1$ kV/cm.

and a 3-kG magnetic field applied perpendicular to the low frequency electrical field. A waiting period of several minutes was often required but there was no indication that a 1-mm separation was an upper limit for the formation of domains.

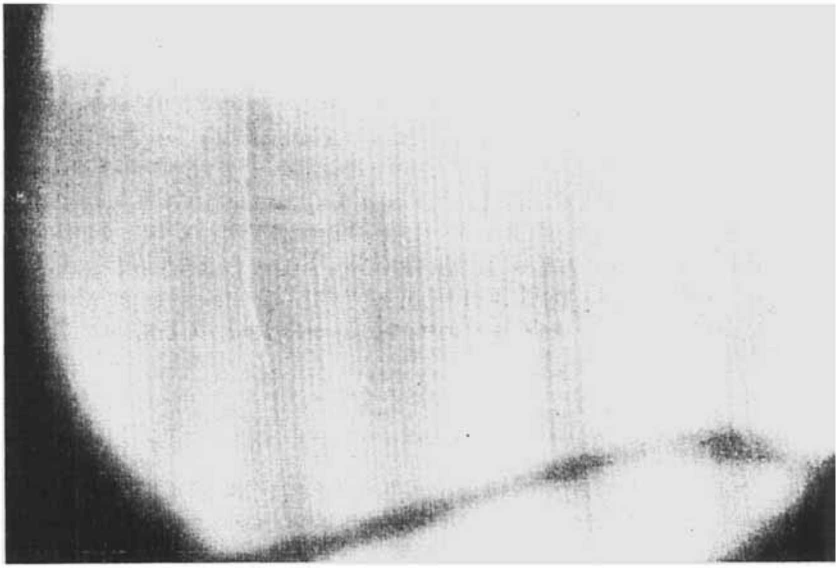
Figure 3 illustrates the size of flow cells at 100–200 V/cm above threshold. A small amount of dye was placed on each of the conducting plates before the cell was filled with sample. Across the top of each photograph can be seen one of the electrodes and the location of the other electrode is near the bottom of the photographs (separation of 2 mm). The photographs were obtained a few seconds after applying the field which allowed enough time for the dye to be pulled into the sample by the material flow. If too much time elapsed the dye mixed with the sample and the contrast was lost. The photographs do not show much contrast but there are five flow lines in each cell indicative of the dye running from one plate to the other. The spacing of these lines is approximately one-fourth the plate separation. When the cell was filled with sample there was a tendency for excess dye to accumulate at the surface near the plates. The effects of this excess dye on the surface can be seen as lines crossing the five flow lines and two curved lines which interfere with one of the flow lines in Figure 3a. This excess dye is easily affected by the threshold field as the observer increases the field to 100–200 V/cm above threshold. There is a very faint line due to a bent cross-hair in the microscope running across the flow lines. The values of the magnetic fields were 4 kG and 16 kG in Figures 3a and b respectively and were applied perpendicular to the electric field. The threshold fields for the flow cells shown in Figures 3a and b were approximately 1 kV/cm and 4 kV/cm respectively. The width of the flow cells are approximately 0.5 mm which is about one-fourth the plate separation. Photographs of this width were much easier to obtain than for wider cells. These results imply that the widths of flow cells depend more on how far the observer is above the threshold field than on the magnitudes of the electric and magnetic field.

B Flow Cells Above Threshold

At electric field intensities much above threshold, dyes tend to mix with the sample making it difficult to obtain photographs. Since there is a variation in the velocity of the fluid when flow cells are created, there is a very small variation in the scattered light which can be photographed. The results from these photographs for various values of the external electric and magnetic fields are presented in Figure 4. Figure 4 shows the flow cell width as a function of an externally applied 50-Hz electric field for various values of a magnetic field applied perpendicular to the electric field. These results are



(a)



(b)

FIGURE 3 Surface of the sample after depositing a dye on the conducting plates and applying a 50-Hz electric field. A magnetic field was applied perpendicular to an electric field which was 100–200 V/cm above threshold.

a. B = 4 kG. Threshold field = 1 kV/cm.
b. B = 16 kG. Threshold field = 4 kV/cm.

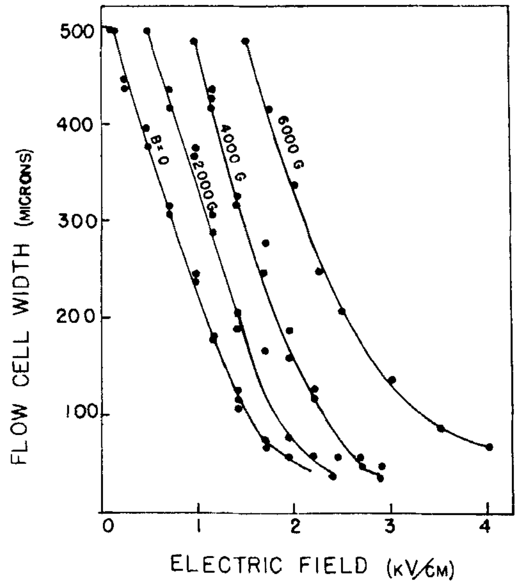


FIGURE 4 Flow cell width as a function of a 50-Hz electric field for various values of a magnetic field applied perpendicular to the electric field. Plate separation =2 mm. Data obtained from photographs of surface with variations in the scattered light due to variations in fluid velocity.

consistent with those using dyes in that the flow cell widths at fields of 100–200 V/cm above threshold are approximately 0.5 mm wide (one-fourth the plate separation). For magnetic fields up to 6000 G and in zero field the width of the flow cells decreases with an increase in the electric field intensity. There was some variation in flow cell widths on any given photograph, but the results show in Figure 4 represent what we believe to be an average width.

A curve at 8 kG which appeared to be consistent with those in Figure 4 was obtained, but the results were difficult to obtain and there were too many uncertainties in the curve to present here. Alignment studies employing dielectric loss measurements as a probe have shown that the aligning mechanism due to the conduction anisotropy is a well behaved process in some samples of MBBA up to at least 20 kG. It appears that an improvement in the experimental techniques is needed for studying flow patterns above 8 kG. The results shown in Figure 4 also indicate a need for improved techniques for investigating flow cell widths below 100 microns. Below approximately 100 microns, the variation of flow cell widths from the average increases rapidly. This behavior might eventually lead to the dynamic scattering mode.

Figure 5 shows the flow cell width as a function of frequency for a 3000 V/cm electric field applied perpendicular to a 4-kG magnetic field. These

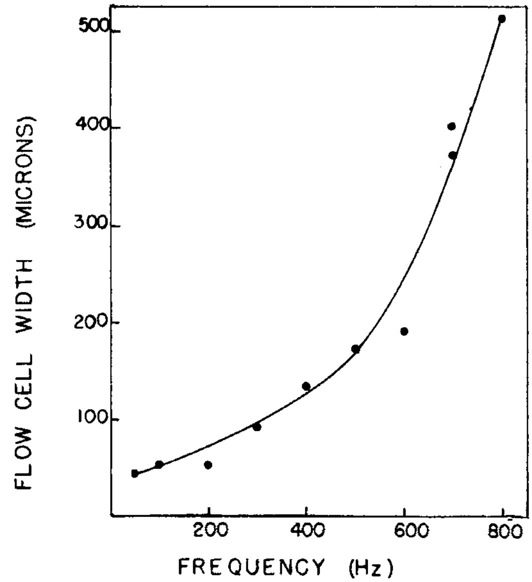


FIGURE 5 Flow cell width as a function of frequency for a 3 kV/cm electric field and a 4-kG magnetic field applied perpendicular to the electric field. $f_c \simeq 900$ Hz. Plate separation $= 2$ mm.

results show that the flow cell width increases with frequency up to the cutoff frequency above which flow patterns are not observed. This is consistent with the formation of Williams domains in that the threshold voltage becomes very high as the cutoff frequency is approached.

C Average State of Alignment

Measurements of the microwave dielectric loss are used to provide some information about the average state of alignment and the results are shown in Figure 6. The general behavior is similar to that reported for other nematic materials in the conduction regime. When the director is normal to the microwave electric field (as indicated on Fig. 6) the state of alignment corresponds to the alignment in the visual cell (used for visual observations) with a magnetic field of 2 kG or higher and an electric field perpendicular to the magnetic field but below threshold values. The dotted line $(\varepsilon_{\parallel}'' + \varepsilon_{\perp}'')/2$ represents an ordering such that the average angle of rotation for the directors is 45 degrees with respect to the initial alignment.

Figures 4 and 6 show that the thresholds for alignment due to ionic conduction and the formation of flow cells are comparable. Different samples of MBBA were used for the results shown in Figures 4 and 6 so very small

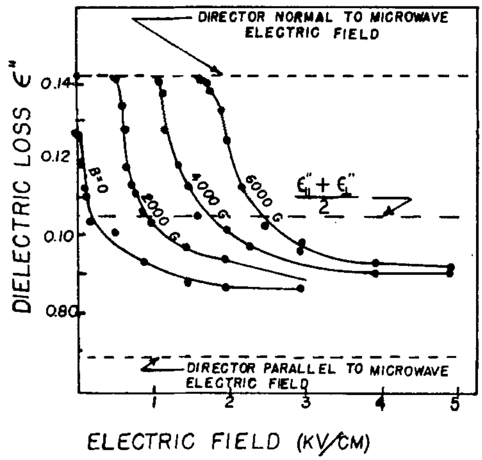


FIGURE 6 Dielectric loss at a microwave frequency of 24 GHz as a function of an externally applied 50-Hz electric field. The individual curves are for various values of a static magnetic field applied perpendicular to the electric field.

variations can be expected. For $B = 0$, the flow cell widths change very little from near threshold to the state of alignment corresponding to a dielectric loss $\varepsilon'' = (\varepsilon''_{\parallel} + \varepsilon''_{\perp})/2$. For a 2000 G field the flow cell width changes appreciable and for 4 and 6-kG fields it reduces by a factor of about two for a 45 degree rotation of the directors. The small value of electric field intensity for $B = 0$ and a state of alignment corresponding to $\varepsilon'' = (\varepsilon''_{\parallel} + \varepsilon''_{\perp})/2$ suggests that the torque due to the distortion of the orientation pattern is small compared to that due to the electric and magnetic fields. The near even spacing of the curves for 2, 4, and 6 kG in Figure 6 also implies a small torque due to the distortion of the orientation pattern, but for flow cell widths below 100 microns this torque can become much more effective.

CONCLUSION

Although quantitative results have been presented to provide information about flow cells in bulk samples of a nematic material, great care must be taken in the interpretation of the results. These results provide information about general behavior in bulk samples but there are variations if the dimensions of the sample cells are changed or the samples become impure before a set of data is complete. If a sample of MBBA is open to humid weather for a few days, the results can change appreciably. We have observed that if the separation of the plates is reduced, the width of the flow cells corresponding to given field intensities is considerably reduced.

All the samples used in this work were in cells with depths of approximately 2 cm or more but most of the information was obtained by observing the surface. However, when a dye was placed on the conducting plates before applying the field, a change in focus of the microscope did indicate that the dye was also pulled into the sample below the surface, and the motion of small particles that were added to the sample indicated that the motion of the fluid was primarily parallel to the electric field and there certainly was some motion below the surface. The motion much below the surface still has to be investigated. However, the application of the electric and magnetic fields perpendicular to each other and parallel to the surface may simplify the behavior. In the absence of a magnetic field the motion may be more complicated.

At electric field intensities just above threshold, the flow cells usually extend from one electrode to the other, but at high intensities they often appear to be shorter and are generated and dissipated rapidly. Because of boundaries and rapid motion of the fluid, there is turbulence which makes it more difficult to investigate narrow flow cells. Experiments involving NMR[6,7] and EPR[8] in nematics, where the relative effectiveness of the electric field was much greater than that due to the magnetic field in producing alignment, have indicated that a large fraction of the molecules align at small angles with respect to the electric field. In view of these results one should probably consider an ordering similar to that illustrated in Figure 1, but having the trace of the director look more like a saw-tooth wave. In this case, the preferred direction for most of the molecules would form a small angle with respect to the electric field.

Some preliminary observations were made with an external magnetic field parallel to a 50-Hz electric field. These results indicated that there were flow cells with widths comparable to those reported in Figure 4, and that the flow cell width decreased with an increase in the electric field intensity. The preliminary measurements indicated that this investigation would be more difficult than the work reported here, but we believe that it should be seriously considered in future investigations.

Although MBBA was adequate for the work reported here, we believe that other materials should be considered for future investigations. For future studies it is likely that materials will be needed that can be doped to change their dielectric and conductivity anisotropies without producing appreciable changes in other properties.

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