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Flow Patterns and Molecular Alignment in a Nematic Liquid Crystal due to Electric Fields

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A mechanism exists for producing molecular alignment in liquid crystals which is associated with ionic conduction anisotropy and hydrodynamic flow. This flow can be investigated with the help of either dyes, dust particles, or scattered light. Previous work on MBBA, which employed magnetic fields up to 6 kG, showed that flow cells usually extended from one electrode to the other and exhibited a domain width dependent on the strengths of the applied electric and magnetic fields. This work has been extended to include magnetic fields up to 16 kG. The thickness of the samples were 0.2 cm and 100 microns. The effect of the dielectric anisotropy on the flow cell width has also been investigated. Data which relates the flow cell width to the average state of alignment will also be presented.

INTRODUCTION

Flow patterns due to electric fields have been observed[1] recently in bulk samples of a nematic liquid crystal. The experimental setup for these observations is shown in Figure 1. The results were obtained from photographs of the surface of the sample while applying external electric and magnetic fields. The 50 Hz electric field (conduction regime) was applied to the electrodes which established an electric field perpendicular to the magnetic field. For low values of the electric field, dyes were used to show the existence of flow patterns. This was accomplished by placing a thread of dye on the surface of the sample or painting the electrodes with a dye before filling the cell and observing the movement of the dye when the electric field was applied. The flow cells usually extended from one electrode to the other and exhibited a domain width dependent on the strengths of the applied electric and magnetic fields.

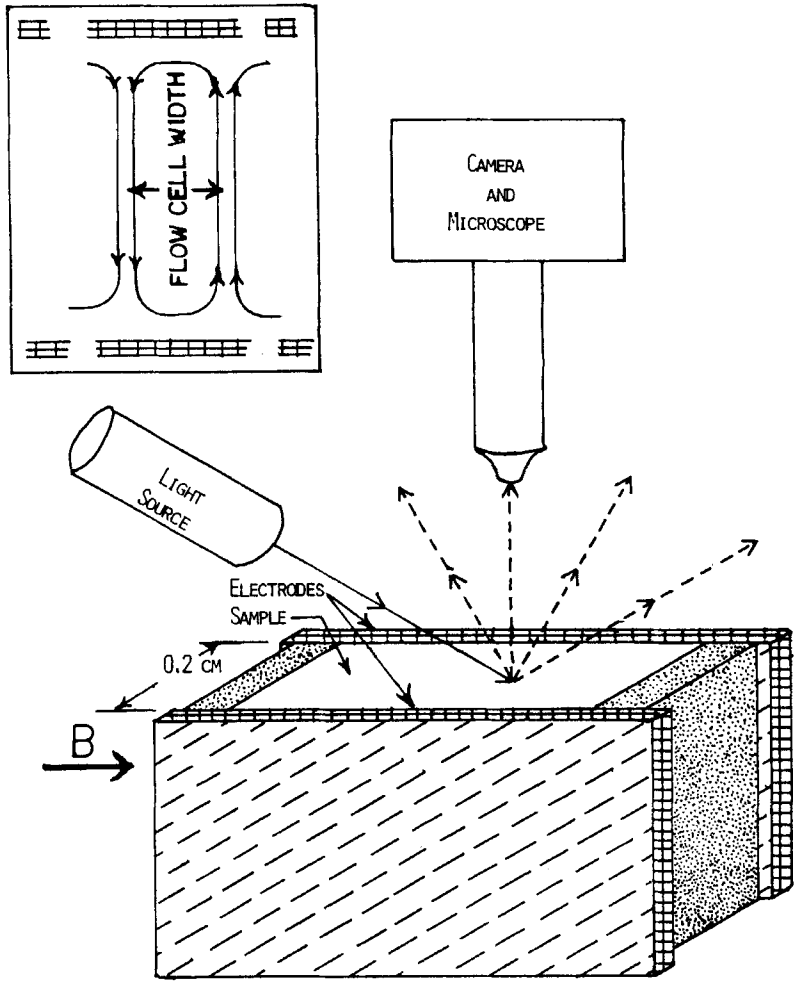

FIGURE 1 Schematic diagram for optical observations.

For low values of the electric field intensity, dyes worked reasonably well, but at higher fields with rapid motion and narrow flow cells, they mixed so quickly with the sample that photographs were difficult to obtain. Since there is a variation in the velocity of the fluid when flow cells are created, there are small variations in the scattered light which can be photographed. Results from these photographs were reported for magnetic fields up to 6 kG and electric fields up to a few kV/cm. We reported that it was difficult to obtain results for magnetic fields above 6 kG. Recently, we have been able to obtain results up to 16 kG and could probably go higher. The objectives

of the work presented here, are to extend the earlier work[1] to much higher field strengths, to investigate the effect of the dielectric anisotropy, and to point out that flow cells similar to those reported here can also be observed in samples approximately 100 microns thick.

Although the experimental techniques employed for the work reported here are different from those reported by other workers,[2] the results near threshold fields are similar to the results obtained by others, but at higher fields the state of alignment appears to be different from much of the earlier work. We suggested earlier[1] that the direction of the nematic director may look more like a saw-tooth wave than a sine wave when the relative effectiveness of the electric field (conduction regime) for producing molecular alignment is greater than that of the magnetic field.

The material used earlier[1] and in the present work was *n*-(*p*-methoxybenzylidene)-*p*-butylaniline (MBBA) with a resistivity of approximately 10^9 ohm-cm and the work was carried out at room temperature. The frequency of the electric field was chosen so that the mechanism primarily responsible for the alignment of the director was associated with the ionic conductivity anisotropy, and that a variation by a factor of at least two in frequency would not be noticeable. MBBA was not necessarily an ideal material for this work, because it tends to absorb water which can result in changes in its properties. It was a convenient material to investigate because the work could be carried out at room temperature, and a considerable amount of information is available on this material. Since the primary objective was to look for general behavior, MBBA appeared to be adequate for this work. The experimental techniques used in obtaining flow cell widths for the work reported here are the same as employed earlier[1] and are shown in Figure 1.

RESULTS AND DISCUSSIONS

A Flow cells in MBBA.

Figure 2 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. The flow cells usually extended from one electrode to the other which was a distance of 0.2 cm. The results show that the flow cell width decreases more rapidly in the presence of low magnetic fields than in high fields for comparable changes in the electric field intensity. Data for these results were often difficult to obtain, particularly, for high values of the magnetic field. There was more contrast at low fields, but the patterns were more uneven. The data were obtained by waiting until a reasonably well

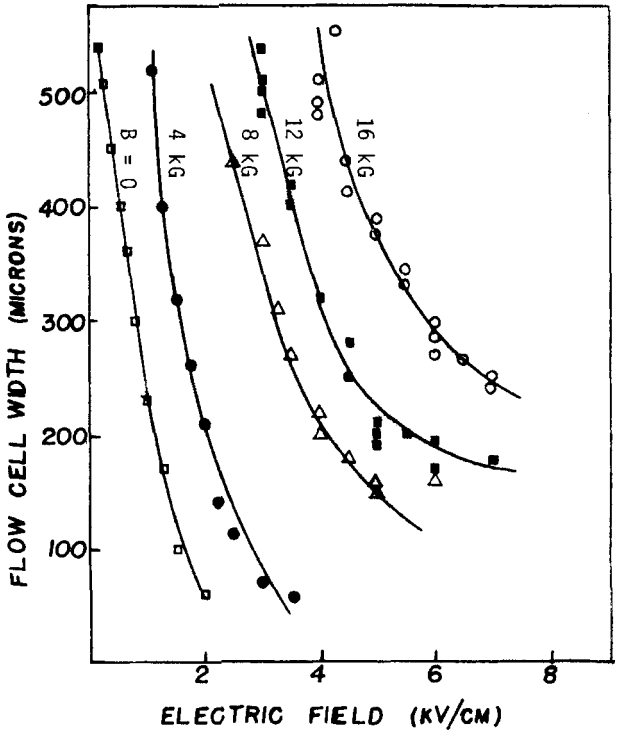

FIGURE 2 Flow cell width for MBBA as a function of a 50 Hz electric field for various values of a magnetic field applied perpendicular to the electric field. Plate separation = 0.2 cm.

spaced pattern appeared, and we assumed that this pattern represented the most probable flow cell width under the given conditions. The life time of these even patterns became very short for high electric field strengths. In low fields, a considerable amount of time was also required for observing most of the reasonably well spaced flow patterns. The mechanism responsible for the flow patterns will be discussed in the conclusion.

B Flow patterns and dielectric anisotropy.

MBBA was doped with small amounts of *p*-[(*p*-methoxybenzylidene)-amino]benzonitrile (PMBAB) to change its dielectric anisotropy. PMBAB has a very large positive dielectric anisotropy ($\Delta\varepsilon' \sim +15$) while MBBA has a dielectric anisotropy $\Delta\varepsilon' = -0.56$ at 25°C. Figure 3 shows the flow cell width in a sample with a dielectric anisotropy $\Delta\varepsilon' \sim 0$. Alignment studies indicated that the dielectric anisotropy may have been slightly positive.

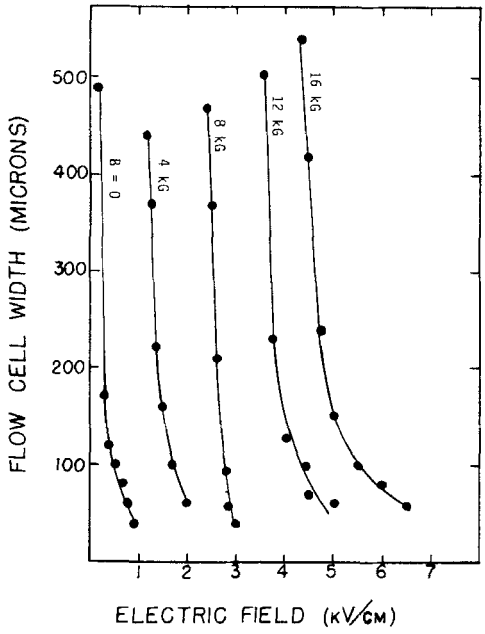

FIGURE 3 Flow cell width for doped MBBA ($\Delta\varepsilon' \sim 0$) as a function of a 50 Hz electric field for various values of a magnetic field applied perpendicular to the electric field. Plate separation = 0.2 cm.

A comparison of Figures 2 and 3 shows that the dielectric anisotropy does have an effect on how rapidly the flow cell width decreases with an increase in electric field intensity. The flow cell width decreases more rapidly with an increase in electric field intensity as the magnitude of the dielectric anisotropy decreases. Results for dielectric anisotropies of $\Delta\varepsilon' = -0.14$ and -0.40 support this trend.

Attempts were made to measure the flow cell widths for samples exhibiting positive dielectric anisotropies, but when the anisotropy was appreciably positive no characteristic flow cell patterns were observed. There was fluid motion and an occasional flow cell, but a reasonably well spaced pattern was not observed. In *p*-azoxyanisole, which had been doped with PMBAB, the aligning mechanism that is associated with the conductivity anisotropy is effective for dielectric anisotropies up to approximately $\Delta\varepsilon' = +1$. This suggests that further studies on materials exhibiting positive dielectric anisotropies might be worthwhile. The flow cell width may change so much with small changes in electric field intensity that reasonably well spaced flow cells are unlikely, but probably flow cells of various sizes are present. With the experimental setup employed in this work it would be difficult to distinguish between turbulence and a system of mixed modes.

The results for $\Delta\varepsilon' \sim 0$ will be further discussed after we have presented the results on the average state of alignment.

C Average state of alignment.

Measurements of the microwave dielectric loss can be used to provide some information about the average state of alignment and the results for $\Delta\varepsilon' \sim 0$ are shown in Figure 4. When the director is normal to the microwave electric field (as indicated in Figure 4) the state of alignment corresponds to the alignment in the cell for visual observations with a magnetic field of 2 kG or higher applied parallel to the electrodes and no electric field. The dotted line $(\varepsilon''_{\parallel} + \varepsilon''_{\perp})/2$ represents an ordering of the molecules such that the average angle of rotation of the director is 45° with respect to the initial alignment.

Figures 3 and 4 show that the thresholds for alignment changes due to ionic conduction and the formation of flow cells are comparable. Since the torque associated with the dielectric anisotropy in MBBA is in the same direction as that of the magnetic field we might expect the threshold for zero dielectric anisotropy to be appreciably higher than that for a dielectric anisotropy of $\Delta\varepsilon' = -0.56$. The ratio of the conductivity parallel to the director

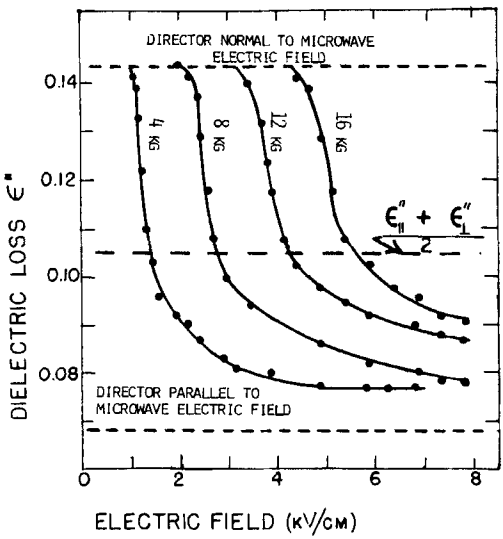

FIGURE 4 Dielectric loss at a microwave frequency of 24 GHz as a function of an externally applied 50 Hz electric field. The individual curves are for various values of a magnetic field applied perpendicular to the electric field. The magnitude of the dielectric loss is a measure of the average state of alignment.

to that perpendicular was slightly larger than 1.5 for undoped MBBA but in the doped sample it was below 1.5, which should account for comparable thresholds in the two cases. Although it was pointed out that MBBA is not an ideal material for quantitative investigations, Figures 3 and 4 provide a good qualitative picture indicating how the flow cell width can be related to the average state of alignment.

A comparison of the results in Figure 4 with those in Figure 6 of an earlier publication[1] shows that there is a greater rotation of the director in the doped MBBA than in pure MBBA for comparable changes in the electric field intensity. This appears to be consistent with the relationship for the torques involving the conductivity and dielectric anisotropies as stated by Helfrich.[2] Since the average state of alignment changes more rapidly in doped MBBA ($\Delta\varepsilon \sim 0$) than in pure MBBA for comparable increases in the electric field intensity, this may be a partial explanation for the rapid decrease in flow cell size in doped MBBA. However, it does not appear to be adequate to explain such a rapid change. It will be difficult to further explain this rapid decrease until measurements are available on other materials or more information is available on the effect of the dopant on the properties of MBBA.

CONCLUSIONS

An interesting aspect of the work reported here is that some of the results can be extended to include thin samples which exhibit dynamic scattering. Lim and Margerum[3] have recently investigated dopant effects on dynamic scattering and have shown evidence of material flow between the electrodes which is associated with the dynamic scattering mode. Although the techniques are different from those discussed here, their work did suggest that some of the work on bulk samples could be extended to thin samples. We have made some preliminary observations of flow patterns in samples approximately 100 microns thick. When observed parallel to the electrodes, flow cells similar to those reported in bulk samples can be observed which decrease in width as the voltage is increased. If we look perpendicular to the electrodes we can see dynamic scattering. At the present time, we do not understand the relationship between these results and those of Greubel and Wolff[4] who investigated electrically controllable domains in much thinner samples (3–10 microns). The results are probably related to some of the work mentioned earlier.[2] Our observations do not explain the actual arrangement of molecules that scatter the light in the dynamic scattering mode. Although work on thin samples appears to be more interesting to most

investigators, bulk samples do have some advantages for much of this investigation. We are able to use a much wider range of fields (lower fields) and we may be able to reduce some of the wall effects.

There are still many experimental details concerning the work discussed in this article that need to be further investigated, but the results appear to give a reasonably good qualitative picture of molecular alignment and material flow. We have assumed that we can explain the behavior in the bulk sample from observations on the surface. This may be a reasonable assumption when magnetic and electric fields are applied perpendicular to each other and parallel to the surface, because the fields could tend to reduce the motion from three to two dimensions. We also need to know more about the mechanism that is responsible for scattering the light from the surface. Much of this scattered light appears to be due to defects which are created by the flow, and many of these defects appear to be disclination lines on the surface. Dust particles would often move with their greatest velocity very close to the lines but not on the lines. This seems to imply that defects are created where the velocity is a maximum. The defects could even be attached to the wall and the fluid could move while the defect remains relatively stationary. These defects can be stretched and bent by the flow of material making it difficult to obtain a photograph which indicates an even spacing of the flow cells. The actual flow patterns in the fluid may be much better spaced than indicated from observations of the surface. Although we are suggesting that defects due to material flow may be responsible for much of the scattered light from the free surface, we do not intend to imply that the light scattered through the electrodes (usually referred to as the dynamic scattering mode) is necessarily due to these effects. Chang[2] has suggested that the onset of the "dynamic scattering" state from a microstructural point of view, may take place principally by the motion of point and line disclinations.

From a theoretical point of view, we probably need to investigate the possibility of a state of alignment where the director can be represented by a saw-tooth wave. When the results presented in this paper are related to those from magnetic resonance experiments,[5] they suggest the possibility of this type of distribution. This applies[1] when the aligning mechanism which is associated with the conductivity anisotropy is more effective than that due to the magnetic field. A detailed analysis would have to be concerned with wall effects if an attempt is made to relate flow cell width to particular values of electric and magnetic field strengths. We have observed that if the plate separation is reduced, the flow cell width is also reduced for a given value of the electric field intensity. The results shown in Figure 4 could be related to those in Figure 3 because the dimensions of the cells were the same in both experiments. More data are needed concerning the

affect of walls on the flow cell width. A theoretical treatment would probably be easier if more results were available when the dielectric anisotropy was zero because the mathematics should be simpler.

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