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

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Version of record first published: 28 Mar 2007.

To cite this article: E. F. Carr , P. H. Ackroyd & J. K. Newell (1977): Dynamic Scattering and Material Flow in a Nematic Liquid Crystal, Molecular Crystals and Liquid Crystals, 43:1-2, 93-101

To link to this article: http://dx.doi.org/10.1080/00268947708084938

Mol. Cryst. Liq. Cryst., 1977, Vol. 43, pp. 93–101

Printed in Holland

Dynamic Scattering and Material Flow in a Nematic Liquid Crystal

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(*Received November 16, 1976; in final form April 1, 1977*)

Results are presented which show how defects in a nematic liquid crystal can be used to investigate material flow when external electric fields are applied. These results include measurements of the size of flow cells, which were created between the electrodes by an electric field. The electrodes, which were made of transparent conductive coated glass, were placed in a vertical position so that flow cells could be observed from the top while dynamic scattering was observed when viewing normal to the electrodes.

INTRODUCTION

Material flow due to electric fields has been investigated[1] recently in bulk samples of a liquid crystal. This involved measurements of the size of flow cells which were created, between the electrodes, by ac electric fields (conduction regime). The flow cells usually extended from one electrode to the other and their size depended on the magnitudes of externally applied electric and magnetic fields. This work was extended[2] to include magnetic fields up to 16 kG and some preliminary observations on flow patterns in thin samples (approximately 100 microns thick). The results were obtained from photographs of a surface of the sample between the electrodes while external electric and magnetic fields were applied. Since there was a variation in the velocity of the fluid within a flow cell, there were small variations in the scattered light which could be photographed. Results are presented here which extend these measurements on the sizes of flow cells to thin samples (135 microns thick) where dynamic scattering was also observed.

The degree of contrast in the photographs for the earlier work[1,2] was generally poor which often made the identification of flow cells difficult. We have discovered that photographs of higher contrast can be obtained if the sample is well aligned just before applying the electric field. These

photographs provide some information about the mechanism which is responsible for the contrast in the scattered light. Photographs in which flow patterns can be easily identified will be used to illustrate these effects.

EXPERIMENTAL

The experimental set-up was similar to that discussed earlier[1,2] except for the method of lighting the surface of the sample. An unpolarized laser beam (approximately 1 mw) was directed parallel to and about 0.3 cm below the surface of the sample. The light which was scattered by the liquid crystal in a direction perpendicular to the beam was sufficient for observation of the phenomena. Since at least one of the electrodes was made of transparent conductive coated glass, the laser beam could be directed perpendicular to the electrodes. It seemed that our best results were obtained when the laser beam was at one edge of the microscope's field of view. The material used in this investigation was a nematic mixture which was purchased from Eastman Organic Chemicals (Cat. No. 11643) and all measurements were made at room temperature. The dielectric anisotropy $\Delta\varepsilon' = -1.6$ and the ratio of the conductivities parallel and perpendicular to the director $\sigma_{\parallel}/\sigma_{\perp} = 2.0$. The resistivity was approximately 10^9 ohm-cm.

MATERIAL FLOW IN BULK SAMPLES

Two photographs (taken in sequence) which indicated flow patterns are shown in Figure 1. The first photograph (Figure 1a) was taken several seconds after a 4 kG magnetic field (applied perpendicular to E) was quickly removed while a 50 volt ac electric field was continuously applied. Since the state of alignment normally changes very slowly in bulk samples, the magnetic field can normally be turned off several seconds or longer before applying the electric field without affecting the results appreciably. For these photographs the electric field was turned on before removing the magnetic field. The 50 volt source did not affect the molecular alignment in the presence of the 4 kG magnetic field. The electrode separation was 0.15 cm and the frequency of the electric field was 50 Hz. Defects were created which became more pronounced with time. The difference between the two photographs is probably associated with a change in the state of alignment. These defects can be seen as evenly spaced lines extending from one electrode to the other. They appeared to move out from the walls and eventually extended to the other electrode. The defects can be used to define flow cells as illustrated in Figure 2.

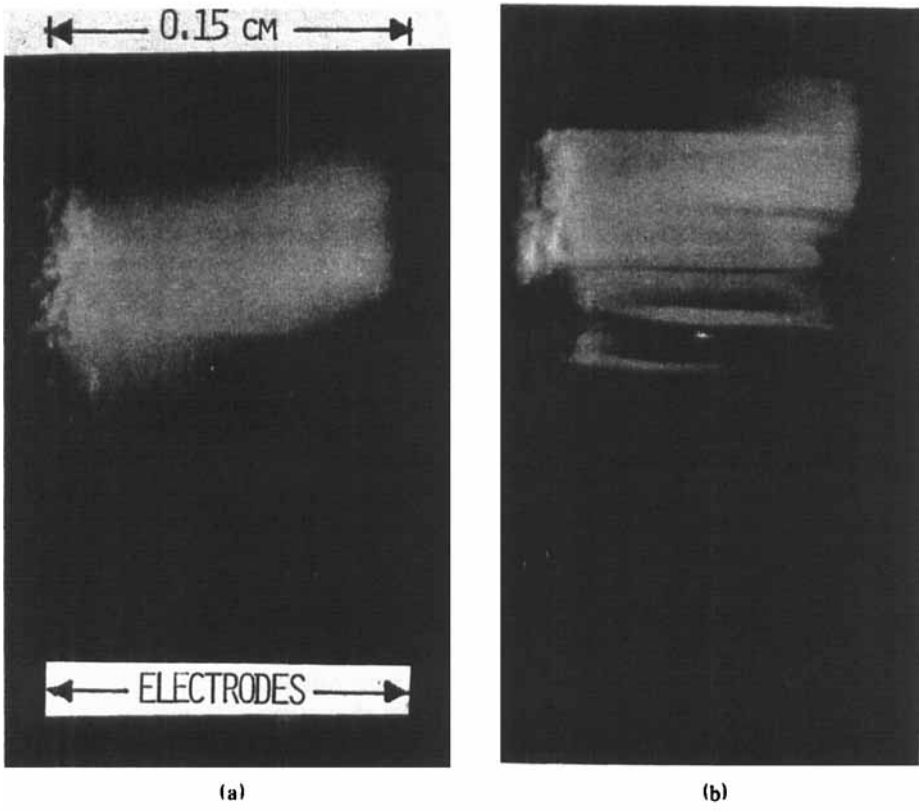

FIGURE 1 Flow patterns due to a 50 Hz electric field at the air to liquid crystal interface. A 50 V source is applied to the electrodes (separation = 0.15 cm). (a) Several seconds after removal of high magnetic field (B parallel to electrodes); (b) Approximately 20 seconds after (a).

The photographs shown in Figure 2 were taken at intervals of approximately 10 seconds. Figure 2a shows a photograph of a defect in the presence of a 4 G magnetic field applied parallel to the electrodes. Changing the magnitude of the magnetic field did not affect the defect appreciably. The defect appeared to get slightly narrower as the field was increased. Defects similar to that shown in Figure 2a have been discussed elsewhere.[3] The second photograph (Figure 2b) was taken after the 4 kG field was removed while a 50 volt ac electric field was continuously applied. The defect (Figure 2a) started to bend when the electric field was applied (Figure 2b) and flow was created. Figure 2c also shows defects similar to those shown in Figure 1, and the distortion of the original defect (Figure 2a) indicates that the magnitude of the velocity of the fluid is a maximum at the defects which were created by the field. Figure 2c also shows that the direction of flow is opposite

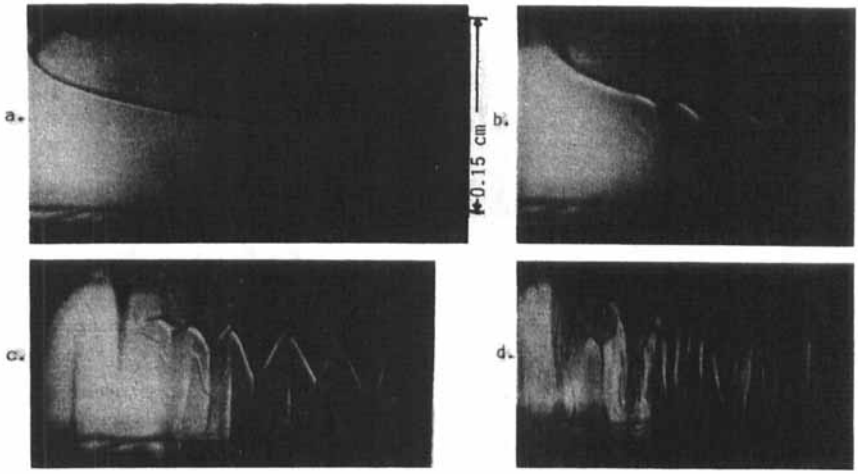

FIGURE 2 Patterns at air to liquid crystal interface. Electrode separation = 0.15 cm. (a) A 4 kG magnetic field applied parallel to the electrodes, and no electric field; (b) Several seconds after removal of high magnetic field with a 50 volt ac electric field applied; (c) and (d) Photographs in (b), (c) and (d) were taken at intervals of approximately 10 seconds.

at adjacent defects. The distance between adjacent defects indicates the flow cell width. Figure 2c also shows another defect that is much thinner than the defect shown in Figure 2a. The bending of this thin defect also indicates the direction of material flow. Figure 2d shows how a defect can be bent making the identification of flow patterns difficult. The type of data shown in Figure 2 can be used to provide information about the velocity of the fluid at various points in the flow cell. We need to only measure accurately the time between photographs to compute average velocities. The maximum velocity (Figure 2) of the fluid under these conditions was approximately 1 mm per minute if we assume that the defect (Figure 2a) moved with the fluid. Observations of floating particles indicated that this was the case.

Flow patterns similar to those in Figure 1 became distorted if the field was left on for a minute or more. If the electric field was removed these patterns often remained for a few minutes. This is to be expected because a sample which has been aligned with a magnetic field will not show much change in the average state of alignment after the field has been off for a short period. For bulk samples this period may be a minute or often much longer. However, this was not the case if there were large temperature gradients. The defects tended to disappear with time after the fields had been removed. Discussions of defects in liquid crystals can be found in the literature.[4]

The exact nature of the defects shown in Figure 1 is not known, but recent work[3] involving domains due to magnetic fields has indicated that the defects may be twist walls. Patterns very similar to those in Figure 1 were obtained[3] when a 1 kG magnetic field ($E = 0$) was applied perpendicular to the electrodes after the nematic director had been initially aligned parallel to the electrodes. When viewing perpendicular to the electrodes, patterns were also observed with a spacing comparable to that observed at the air-to-liquid crystal interface. This implies that, after the magnetic field had been on for about 1 min, the patterns may have been due to inversion walls. The orientation of the nematic director at the defect may be similar to that expected[5] for splay-bend or twist walls parallel to the field.

Patterns were observed when viewing perpendicular to the electrodes using a 60 Hz electric field. The spacing was comparable to that observed at the air-to-liquid crystal interface. This implies, that after a 50 volt electric field had been on for about 0.5 minutes the patterns may have been due to inversion walls. The movement of fluid, as illustrated in Figure 2, would tend to favor a splay-bend wall but the twist wall is more likely to be expected from energy considerations.[4] However, when a magnetic field is simultaneously applied parallel to the electrodes, the orientation of the nematic director may tend to form a splay-bend wall.

Figure 3 shows the flow cell width as a function of the electric field intensity for flow cells of the type shown in Figure 1. The flow cell width is defined as the separation between adjacent defects. We waited approximately a minute for flow cells to form at the lower electric fields, but at the higher fields they

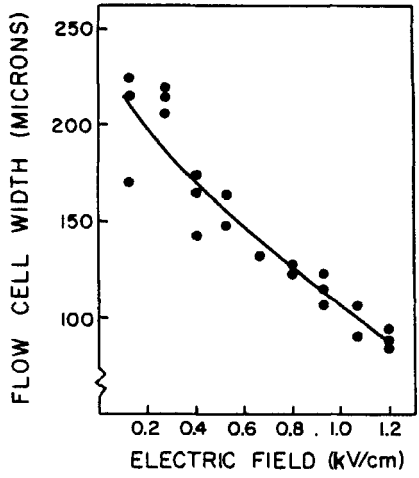

FIGURE 3 Flow cell width as a function of a 50 Hz electric field. Electrode separation = 0.15 cm. The data were obtained from photographs similar to those shown in Figure 1.

formed within a second. The results shown in Figure 3 show that the flow cell width decreases with an increase in electric field intensity which is consistent with earlier results[1,2] using MBBA. The important difference between the work reported here and the earlier work is that in this case we are applying the electric field to a well aligned sample. In the earlier work[1,2] it was necessary to wait until a reasonably well spaced pattern appeared, and it was assumed that this pattern represented the most probable flow cell width under the given conditions. The work presented here shows that for a given field strength the normal situation is not a system of mixed modes if one starts with a well aligned sample. The fact that a reasonably well spaced pattern was not continuously observed when using the earlier method[1] does not necessarily mean that the situation was a system of mixed modes. Defects are not only being created and destroyed when the field is on continuously but they are also being bent and twisted making the identification of flow cells difficult.

Results, which were similar to those in Figure 3, were obtained using the method described earlier[1,2] with the continuously applied electric field. These results showed a flow cell width that was a little narrower than those shown in Figure 3. Results when using *p*-azoxyanisole also showed that the flow cells were larger if one starts with a well aligned sample (director parallel to electrodes) and makes the measurements before the pattern becomes distorted. We do not have an explanation for this difference.

MATERIAL FLOW IN THIN SAMPLES

Flow patterns similar to those shown in Figure 1 were observed in thin samples of the nematic mixture with a plate separation of 135 microns. It was more difficult to obtain data for this separation than in the larger samples because of the higher electric and magnetic fields required. The threshold voltage across the much smaller electrode separation limits investigation to high electric field intensities. As was found in the bulk sample, flow patterns were more difficult to obtain at high electric fields.

We were able to use high electric fields by employing the method similar to that reported earlier[1,2] and this method was used to obtain the results shown in Figure 4. The life-time of flow cells that were initiated from well aligned samples was very short in high electric fields.

Figure 4 shows the flow cell width as a function of electric field intensity for a cell with an electrode separation of 135 microns. The lower curve shows the flow cell width with no magnetic field and the upper curve gives results in an 8 kG magnetic field. The 8 kG magnetic field was applied perpendicular to the electric field. The results were similar to those[1] discussed

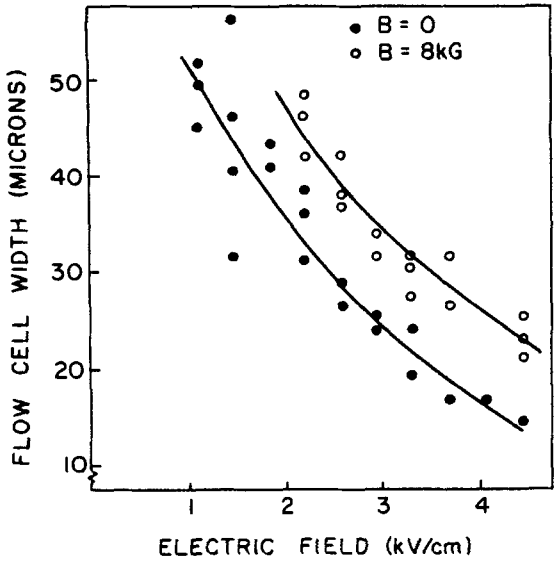

FIGURE 4 Flow cell width as a function of a 50 Hz electric field. Electrode spacing = 135 microns. The results for an 8 kG magnetic field were obtained with the magnetic field applied perpendicular to the electric field.

earlier for bulk samples in that the flow cell width decreased with an increase in electric field intensity, but was larger when a magnetic field was applied perpendicular to the electric field. Defects were observed for fields below 1 kV/cm, but it was difficult to obtain good data with this method at the lower fields. The sample cell for these measurements was made from two pieces of transparent conductive coated (tin-oxide) glass which were separated by a teflon spacer. The pieces of conductive coated glass were in a vertical position so that flow patterns could be observed from the top while dynamic scattering was observed visually when viewing perpendicular to the electrodes. The electrodes were cleaned with acetone and rubbed in a direction parallel to the air-to-liquid crystal interface, but we do not believe that the rubbing had an appreciable effect. The wall effects did not produce either a good homogeneous or homeotropic alignment in this material. These results show that the flow cells, which are similar to those discussed earlier for bulk samples, are associated with the dynamic scattering mode.

Figure 4 shows a flow cell width of approximately 50 microns ($B = 0$) for an electric field intensity of 1 kV/cm, whereas, Figure 3 shows a flow cell width of approximately 100 microns at this field strength. We believe that this difference in flow cell widths is due to the difference in electrode separations. Earlier results[1] also showed that the flow cell widths depended on the electrode separation.

Flow cells that were created from well aligned samples were investigated for an electrode separation of 135 microns. The initial alignment was produced by a high magnetic field. This field, which was applied parallel to the electrodes, was quickly removed while applying the 50 Hz electric field. While both fields were applied the alignment was determined by the magnetic field. The flow cell widths were comparable to those shown in Figure 4. This investigation was more difficult when working at higher electric field strengths, but we were able to identify flow patterns at lower fields. With a potential difference of 7 volts ($E = 520$ V/cm) applied to the electrodes we were able to identify flow patterns with a spacing comparable to the separation of the electrodes but at 6 volts we did not obtain any well spaced patterns. When viewing with a microscope perpendicular to the electrodes Williams domains were observed at 6 volts. Williams domains were observed at 7 volts but they soon started to break up. Although the domains did tend to break up at 7 volts we did not observe any rapid turbulent flow but visual observations indicated an appreciable amount of light scattering with a long rise time. These observations at 6 and 7 volts appear to be consistent with a threshold value of 6.4 volts reported by Eastman Kodak Company.

CONCLUSION

We have provided good evidence that flow cells are associated with the dynamic scattering mode for nematic materials with a sample thickness of 135 microns and it is likely that this applies to much thinner samples. This work may even be related to that of Grenbel and Wolff[6] who investigated the domain width as a function of the applied voltage in sandwich cells with thicknesses of less than 10 microns. They reported that the inverse domain width is proportional to the applied electric field, while "dynamic scattering" does not occur. The work reported here also appears to be consistent with other results[7] in this area. We did not investigate any effects due to wall alignment, but we have made a few observations with other nematic materials. We have used *p*-azoxyanisole and MBBA which exhibits a homogeneous orientation with untreated surfaces, and also a nematic mixture from E. Merck (No. 5A) which has been doped to exhibit a homeotropic orientation. The behavior of all these materials appears to be consistant with what is reported here.

The results discussed here indicate that if the initial condition (before applying an electric field) was a well aligned sample with the nematic director parallel to the electrodes, the flow cells which were created by the electric field do not correspond to a system of mixed modes. This does not necessarily mean that the earlier work[1,2] involved a system of mixed modes.

Although many types of defects are created by the electric field, twist walls may be primarily responsible for the patterns discussed here. When the magnetic and electric fields are applied simultaneously we should consider the possibility of a splay-bend wall. If walls are present the space charge density should be a maximum at the wall because of the conductivity anisotropy. This would tend to favor a triangular distribution for the alignment of the director rather than one that could be represented by a cosine function, which might help explain some of the magnetic resonance experiments.[8] These experiments have indicated that in a large portion of the sample the nematic director aligns at small angles with respect to the electric field. These angles were in the neighborhood of 20°. The forces due to the interaction of the space charge with the electric field, which act on the walls, would tend to shear the sample. It is interesting to note that in the first article on the dynamic scattering mode, Hielmeier, Zanoni and Burton reported that dynamic scattering could be produced by mechanically shearing the sample.

Although we have suggested that flow patterns which may involve inversion walls are associated with the dynamic scattering mode, we do not intend to imply that most of the scattered light is necessarily due to the inversion walls. It has been suggested by Chang[7] and de Gennes[9] that the disclinations may be involved with the dynamic scattering mode. This is not inconsistant with the presence of inversion walls because point and line disclinations can result from the process of creating walls.

Acknowledgement

The authors wish to express their appreciation to Mr. R. W. H. Kozlowski for many helpful discussions.

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