

## CHAPTER III

DISCUSSIONS AND CONCLUSIONSIII.1 General Theoretical Considerations

The general theoretical ideas presented here regarding the electrohydrodynamics of liquid crystals are based primarily on a consideration of the local field effects. These effects presumably determine the motions of the liquid crystals leading to formation of the observed domains. The principal idea is that molecules of a liquid crystal can form clusters of dipoles which are stationary in a uniform electric field but become mechanically unstable in a nonuniform electric field. Evidence from nuclear magnetic resonance suggests that the postulated clusters of molecules should generally exist in the nematic phase of liquid crystals. The results of the present experiments can be very readily understood if it is assumed that dipolar clusters give rise to a nonuniform local field. In the presence of an external electric field, molecules or clusters of molecules of a liquid crystal are polarized, and this induced polarization also contributes to the local field.

The local field at any point in the liquid crystal in the presence of the external field can be divided, as in Kirkwood's statistical theory, into three terms. The first term represents the applied electric field and the electric field from the polarization induced by it without any distortions of the molecules. The second term represents the electric field from induced curvature strains, and the third represents the field from the polarization of neighbouring molecules or molecular clusters by a single dipole or a representative cluster under consideration.

So far, realistic local fields in a liquid crystal have never been calculated because of the difficulties involved. First of all, the molecules of a liquid crystal

are typically polar, having a rod-like shape with large electric dipole moments either perpendicular or parallel to their long axes which also tend to point in the same preferred direction. A cluster can be formed from these molecules in such a way that a net dipole is produced. The effect of the local field on the representative dipole is not only to align it in the local field direction but also to cause it to move along the local field gradient.

Recently<sup>1</sup>, the local field in a simple lattice model of oriented liquid crystals has been calculated, and it has become evident that the anomalous alignment of liquid crystalline molecules may now be reasonably well understood in terms of the local field effects. However, the details of these results are not entirely in agreement with the widely accepted Helfrich theory regarding the anomalous alignment. In particular, it has been found<sup>1</sup> that the dielectric anisotropy changes its sign when the mean dielectric constant exceeds a certain value. This result explains why a liquid crystal with negative dielectric anisotropy is aligned with long axes of the molecules parallel to the field direction. A more realistic calculation of the local field using a radial distribution function of liquid crystalline molecules or clusters would certainly be valuable. Unfortunately, this presents a number of problems and is beyond the purpose of the present research. It should be noted that there are also significant contributions to the local field from dipoles at large distances from the representative dipole in the bulk medium. The problem of quantitative estimation of these contributions in the case of a typical liquid crystal has so far never been solved.

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<sup>1</sup>D.A. Dunmur, "Local Field Effects in Oriented Nematic liquid Crystals.", Chem.Phys.Letters, 10 (1971), 49

Since the representative dipole moves in the presence of an electric field, the friction to the motion must be taken into account. All nematic liquid crystals possess uniaxial properties and thus the viscosity along the preferred axis, which usually directs along the long axes of the molecules, would be lower than that perpendicular to it. The anisotropic viscosity has been studied by Ericksen<sup>2</sup> and Leslie<sup>3</sup>. The domain pattern may change considerably because of the viscosity and this fact has been recognized only recently. Thus the representative dipole or dipolar cluster would tend to move along the preferred direction along which the viscosity is lower.

It is clear that in the presence of an electric field the dipoles or clusters in a liquid crystal are moving and hence the foregoing analysis should be extended to include the effects of the local field resulting from moving dipoles. However, this would present still further difficult problems. Also, it has been evident from the experimental results that in general the motions of the domains tend to become more vigorous and disordered with increasing field strengths. This certainly reflects the corresponding complexity of the local fields.

### III.2 Discussions of Experimental Results and Conclusions

#### III.2.1 Nematic liquid crystals

Almost all earlier theories described the domains in nematic liquid crystals as resulting from static distortion effects, and it had never been properly recognized before the

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<sup>2</sup> J.L. Ericksen, Arch.Ration Mech.Anal., 4 (1960), 231

<sup>3</sup> F.M. Leslie, Quart.J.Mech.Appl.Math., 19 (1966), 357

observation made by Penz<sup>4</sup> that the domains could in fact arise from concerted dynamic effects. According to Penz<sup>4</sup>, the vortical motion of materials in a liquid crystal layer produced a set of cylindrical lenses and these lenses can produce real and virtual images just above and below the liquid crystal layer. The vortical motions and the existence of two sets of the bright domain lines have been confirmed by the present investigations.

The Helfrich theory<sup>5</sup> which was first formulated for the liquid crystals in the presence of a d.c. electric field has recently been extended by the Orsay Liquid Crystal Group<sup>6</sup>. The extended theory has shown that regular domain patterns can possibly be obtained in two important régimes. At low frequencies, the "conducting régime" creates the hydrodynamical cellular flow, whereas at high frequencies, the "dielectric régime" plays a significant role. However, it is not yet clear what effects could make the domains become visible. The present results have indicated that some kind of dynamic effect may be responsible for it. On the other hand, it should be noted that the present experiments were mainly carried out by using 50 Hz a.c. electric fields and d.c. electric fields, and thus it is not clear whether the domains observed result from the "conducting régime" or from the "dielectric régime".

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<sup>4</sup> P.A. Penz, "Voltage-Induced Vorticity and Optical Focusing in Liquid Crystals.", Phys.Rev.Letters, 24 (1971), 1405

<sup>5</sup> W. Helfrich, "Conduction-Induced Alignment of Nematic Liquid Crystals.", J.Chem.Phys., 51 (1969), 4092

<sup>6</sup> Orsay Liquid Crystal Group, "Hydrodynamic Instabilities in Nematic Liquids under a.c. Electric Field.", Phys.Rev.Letters, 25 (1970), 1042

If the detailed motions of the liquid-crystal materials are not being considered, the domains observed in the free-lead-electrode cells are in many respects similar to those observed in the transparent-electrode cells. It has been found that there are two sets of bright domain lines in the sample of  $40\ \mu\text{m}$  thickness in free-lead-electrode cells but there is only one set in a sample of  $20\ \mu\text{m}$  thickness. This indicates that the existence and nature of the domains are strongly dependent on the sample thickness. The domain lines in a free-lead-electrode cell are aligned along the field direction. In contrast, the domains in a transparent-electrode cell are aligned in a plane perpendicular to the field direction.

The results from this and earlier experiments<sup>7</sup> indicate that the domains become more disordered in the presence of a d.c. electric field in a cell with either type of electrodes. This fact may be satisfactorily explained in terms of the local field effects. The electric fields required for the first appearance of domains in free-lead-electrode cells have been found to be considerably larger than those required for transparent-electrode cells because of different electrode configurations. On the other hand the threshold field is insensitive to the sample thickness.

At present it is almost certain that the regular polygons obtained in the transparent-electrode cell at a high field strength results from electrohydrodynamic instability. Earlier experiments<sup>7</sup> have shown that thermo-

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<sup>7</sup> Sauvarop Bualek "Electro-Optical Studies of Nematic Liquid Crystals.", M.Sc. thesis, 1971, Department of Chemistry, Mahidol University, Bangkok, Thailand.

hydrodynamic instability is rather unlikely, for it requires impossibly low viscosity. The local field effects may also be able to explain the formation and movements of the domains in both types of electro-optical cells. However, this requires further and more detailed studies and must await some realistic calculations of the local fields in nematic liquid crystals.

It is not yet clear whether the dark grains found in all nematic liquid crystals before an application of an electric field are in any way related to the clusters of molecular dipoles. However, it would be surprising if such clusters are in fact large enough to be resolved by the microscopes used. On the other hand, the results from the present experiments may be enough to indicate that the dark grains have a close connection with the appearance of the domains.

### III.2.2 Smectic liquid crystals

The present investigation is also concerned with molecular arrangements, textures, and electrohydrodynamics of smectic liquid crystals; and the observation of textural motions within thin layers of these materials is reported for the first time. It has already been described that the long focal conics, in the presence of an electric field, move perpendicular to the field direction towards the boundary of the smectic liquid crystal drop and these motions are coupled with a rotation about the long axes of the focal conics. More careful investigations have shown that the axially-symmetric regions of a fan-shaped texture always move with their apices pointing in the direction of motion. The long rods and bâtonnetes in the supercooled

state have been found to rotate about their long axes and vibrate slowly about the field direction. These motions are quite different from those of the polygonal textures.

At low field strengths the polygons are stationary, while at high fields they burst vigorously into a number of focal conics. These textural motions can be qualitatively explained in terms of a theoretical model of molecular arrangements and growth of domains within smectic liquid crystals. Since homogeneous nucleating centres which can be created from fluctuations of order are rare, the principal nucleating centres should be heterogeneous ones which may be dust particles or impurities. For the case of long rods which are obtained when the smectic liquid crystals are cooled down slowly from the isotropic phase, molecules of the smectic liquid crystals can be initially attached to a nucleating center with their long axes pointing radially. Other molecules can then be attracted to these molecules by Van der Waals forces and electric dipole-dipole interactions. The growth mechanism then continues presumably in a spiral fashion until a critical size is reached. If a screw dislocation occurs during the growth mechanism, then the long rods can be formed. Frequently the long rods also contain other faults which can be expected to arise in the growth process.

The suggested spiral growth mechanism for the long rods can also satisfactorily explain the experimental results that the long rods have rotational motions about their long axes. These motions arise because of the existence of electric dipolar couples in the long rods. The electric field exerts torques on dipolar couples and, as a result, the long rods can rotate about their long

axes. The slight vibration of the rods about the field direction may arise either from thermal fluctuation or from the fact that the torques act on the couple which have dipoles not pointing exactly perpendicular to the long axis of the rod.

The optical properties in crossed polaroids of the polygonal texture, in particular the alternately dark and bright concentric bands, indicate that the molecular arrangements should be periodic alignments of long axes of the molecules along the radial direction of the polygons. The dark bands would correspond to the small regions where the long axes of the molecules point mainly upwards or downwards and thus these region would be the optic axes as in ordinary crystals. When polarized light propagates along the optic axes, the plane of polarization is not rotated and thus a large extinction is observed in crossed polaroids. The bright, concentric bands would correspond to the regions where the long axes of the molecules point in other directions, and this causes less extinction of polarized light. Frequently, two dark crossed arms appear along the radial direction of each polygon. This fact may indicate that there are also windings of long axes of the molecules along the tangential lines of the concentric bands and these windings are in phase with neighbouring bands. These molecular arrangements and the optical properties of the polygonal texture are strikingly similar to those of the polymeric textures known as spherulites.

The molecular arrangements in the polygonal texture as described above may aid in explaining why the polygonal textures are fairly stationary at low electric field strengths. According to the suggested molecular arrangements, the electric dipole moments of the molecules may almost



cancel out. At high field strengths, the torques acting on the dipoles may be large enough to break the molecular arrangements and thus to cause the burst of polygons.

Investigations of the optical properties of focal-conic and fan-shaped textures have led to a proposed model for molecular arrangements different from those of the polygonal textures. In crossed polaroids, the dark region at an apex of the fan-shaped texture changes uniformly to a bright region near the dark boundary, which implies that the molecular orientations change uniformly from the central region to the boundary. It is also quite plausible that the long axes of the molecules could be normal to the plane containing the fan-shaped texture. The dark lines and dark boundaries of the fan-shaped and focal-conic textures would then correspond to the regions where the molecular orientations change abruptly with a large extinction of polarized light. It is thus seen that the textural motions of fan-shaped and focal-conic textures may be satisfactorily explained by basic considerations of the molecular arrangements.