CHAPTER I

INTRODUCTION

I.1 Historical Review

The molecular orientations and motions within a thin layer of nematic liquid crystals under the influence of an electric field have been extensively investigated following the discovery in 1963 of the distinct domain pattern by Williams[1]. It was first suggested that the visible domains might be produced as a result of phase separation of the liquid crystals, which could have been induced by the applied electric field. However, Williams ruled out this possibility since phase separation induced by an electric field has never been directly observed in the case of other materials. For such induced phase separation to be possible, it would require a large diffusion coefficient.

Williams[2] then suggested that the domains might be due to the orientation effects induced by an electric field. These effects would be expected to produce regions having different refractive indices. Substantial orientation effects on the individual electric dipoles of the molecules of a liquid crystal would necessarily require a fairly high electric field strength. On the other hand, at rather low field strengths, substantial orientation effects could effectively be produced by a concerted action of a number of molecules of the liquid crystal.

---

[1] R.Williams, "Liquid Crystals in an Electric Field.", Nature, 199 (1963), 27

[2] R.Williams, "Domains in Liquid Crystals.", J. Chem. Phys., 39 (1963), 384

By analogy with ferroelectric effects, Williams[3] proposed that the ordering effects of a nematic liquid crystal together with wall effects could lead to a net dipole moment. The polarizations were divided into small regions called domains, each being separated from its neighbours by domain walls. An electric field would exert a torque on an individual domain which contains a number of parallel dipoles, and thus the net dipole of the domain would be oriented. Within the thin layers of the domain walls, where the polarizations of neighbouring domains were antiparallel, an electric field would exert no torque. These oriented and unoriented regions could be expected to exhibit two different refractive indices, the difference between which might be large enough to cause the separate domains to become visible.

It has been evident from various experiments[3,4,5] that nematic liquid crystals exhibit several characteristics of ferroelectricity, such as hysteresis loops, switching transient currents, and dielctric anomalies at low frequencies. The transient behaviour has been studied by Heilmeier[5]. His conclusion was that the rise time of the domain was proportional to the square of the sample thickness and varied inversely as the applied voltage. Similar effects have also been found in solid ferroelectrics.

---

[3] R.Williams and G.H.Heilmeier, "Possible Ferroelectric Effects in Liquid Crystals and Related Liquids.", J.Chem.Phys., 44 (1966), 683

[4] G.H.Heilmeier and P.M.Heyman, "Note on Transient Current Measurements in Liquid Crystals and Related Systems.", Phys.Rev. Letters, 18 (1967), 558

[5] G.H.Heilmeier, "Transient Behaviour of Domains in Liquid Crystals.", J.Chem.Phys., 44 (1966), 644

A more fundamental and rigorous theory of anomalous alignment in a liquid crystal based on the concept of field-induced space charges has been developed by Helfrich[6]. This approach makes use of the well-established continuum theory which has been reviewed by Ericksen[7]. The space charges may arise from the anisotropy of both the conductivity and the dielectric constant. The interaction of space charges with the generating electric field causes material flow in opposite directions. This flow is accompanied b, a shear, which, in turn, exerts a torque on the molecules of the liquid crystal. The torque tends to change the direction of the preferred axis and thus to change the orientation pattern. The torque would also arise from dielectric polarization. According to this theory the visible domains may in fact be the periodic distortions of the orientation pattern[6] caused by electric conduction induced by the torques.

A successful extension of the Helfrich theory to an a.c. electric field has been made by the Orsay Liquid Crystal Group[8]. It has been shown that the onset of the hydrodynamic instability in nematic liquid crystals involves many physical effects. In a d.c. electric field, a unipolar charge injection is involved, whereas in an a.c. electric field the domains may be interpreted in terms of the Helfrich theory

---

[6] W.Helfrich, "Conduction-Induced Alignment of Nematic Liquid Crystals.", J.Chem.Phys., 51 (1969), 4092

[7] J.L.Ericksen, Appl.Mech.Rev., 20 (1967), 1029

[8] Orsay Liquid Crystal Group, "Hydrodynamic Instabilities in Nematic Liquids under a.c. Electric Fields.", Phys.Rev.Letters, 25 (1970), 1642

of the anisotropy of conductivity and dielectric constant. At low frequencies, the space charges are modulated and this has been referred to as the "conducting regime". This conducting regime creates a regular pattern of hydrodynamic cellular flow which would correspond to the Williams domains. The space charges would no longer be able to follow the a.c. electric field above a certain cut-off frequency. Only local bendings in the liquid crystal would be modulated at such a high frequency, and this has been called the "dielectric regime". The pattern arising from such a mechanism was reported[8] to be rather different from the "conducting regime" pattern.

Direct experimental evidence which is contrary to earlier reports has been found by De Jeu[9] in a nematic liquid crystal. Liquid crystals with positive dielectric anisotropy should not form Williams domains as predicted by the Helfrich theory. However, it was found that the domains could be formed not only in the nematic phase but also in the isotropic phase of such a material. This disagreement could satisfactorily be interpreted in terms of the Felici model[10] which is valid for all dielectrics. The space charges due to unipolar charge injection lead to cellular hydrodynamic flow, which should correspond to the Williams domains.

---

[9] W.H. De Jeu, C.J.Gerritsma and A.M.Van Boxtel, "Electrohydrodynamic Instabilities in Nematic Liquid Crystals.", Phys.Letters, 34A (1971), 203

[10] H. Felici, Rev.Gen.Elec., 28 (1969), 717

Another effect has been proposed by Meyer[11] as a possible explanation for the formation and stability of the Williams domains. This is based on an interaction analogous to piezoelectric effects in ordinary crystals. An interaction between a liquid crystal and electric field compatible with the symmetry properties would aid in explaining unusual effects in nematic liquid crystals. An extension of Frank's idea has been made according to which splay or polarization can be externally produced by an electric field or by a mechanical stress.

In the presence of splay, the symmetry of the system changes from nonpolar to polar and thus the polarization is induced or vice versa. On the other hand, in polar liquids the molecules are randomly oriented so that the system is nonpolar. An electric field will splay the structure by aligning the permanent dipole moments, whereas the splay will polarize the molecules by acting on shape polarity. These electro-mechanical effects may help to interpret the domains as alternating regions of splay and bending. This mechanism is analogous to the piezoelectric effects in ordinary crystals, but the former involves curvature strains rather than tensile or shear strains.

More recently, Penz[12] has investigated the domains in detail by introducing tracer particles and observing

---

[11] R.B.Meyer, "Piezoelectric Effects in Liquid Crystals.", Phys.Rev. Letters, 22 (1968), 918

[12] P.A.Penz, "Voltage-Induced Vorticity and Optical Focusing in Liquid Crystals.", Phy. Rev. Letters, 24 (1970), 1405

their motions. He suggested that the bright domain lines were real and virtual images of a microscope-lamp source produced by cylindrical liquid-crystal lenses with voltage-dependent focal lengths. He indicated that there were two sets of bright domain lines, one just above the liquid crystal layer, which corresponded to the set of real images, the other set just below the liquid crystal layer, which corresponded to the set of virtual images. The liquid crystal lenses were formed by vortical motions of materials in liquid crystal layers. The directions of the vortical motions were antiparallel in the adjacent domains.

More detailed investigations have been made by Sauvarop Bualek[13] in her recent thesis. Her results support Penz's conjecture. She was also the first to make use of the observations of the Williams domains in various nematic liquid crystals. In the case of an electro-optical cell with transparent electrodes, she has found that at very high electric field strengths, around 10^4 volts/cm, some liquid crystals form a pattern of regular polygons similar to Bénard cells[14] and she has suggested that this pattern could be understood in terms of an electrohydrodynamic analogue of thermohydrodynamic Bénard cells in ordinary liquids. The onset of instabilities in nematic liquids has been interpreted in terms of the Helfrich theory.

---

[13] Sauvarop Bualek, "Electro-Optical Studies of Nematic Liquid Crystals.", M.Sc. Thesis, 1971, Mahidol University, Bangkok, Thailand.

[14] H.Bénard, "Les Tourbillions Cellulaires dans une nappe Liquide Transportant de la Chaleur par Convection en Regime Permanent.", Ann.Chim.Phys., 23 (1901), 62

I.2 Purpose of the Present Experiments

The principal purpose of the present investigation is to reveal the true nature of the molecular alignments and the domain patterns which can be induced in a thin layer of nematic liquid crystals by means of an intense electric field. Attempts have been made to give a detailed theoretical explanation of the experimental results. An essential part of the theory is the application of Kirkwood's and Cole's theory[15] of dielectrics with generalizations to the case of an anisotropic fluid in an a.c. electric field. It is proposed that, in the nematic phase, several molecules can interact fairly strongly to form a cluster in such a way that the individual dipoles of the molecules would add vectorially up to a finite net dipole belonging to the cluster. The electrohydrodynamics of nematic liquid crystals can thus be discussed in terms of the local field effects. It has also been found that some fine dark grains can usually be observed in nematic liquid crystal layers before an electric field is applied. Investigations have been made in order to find out whether there is any possible connection of this with the formation of the Williams domains.

The present experimental studies also include those of electrohydrodynamics and molecular arrangements of smectic

---

[15] N.E. Hill, W.E.Vaughan, A.H.Price and M.Davies, Dielectric Properties and Molecular Behaviour (London & New York : Van Nostrand Co., 1969), pp. 24-33.

liquid crystals. The textural motions of the smectic liquid crystals have been carefully investigated at various electric field strengths from zero to around 10^4 volts/cm. The results from the experiments on electrohydrodynamics are discussed and interpreted in terms of the effects of the local field on the dipoles which exists in smectic materials. Molecular arrangements of the ordinary focal conics, fan-shaped textures, a new kind of focal conics, polygons and long-rod-shaped textures are proposed on the basis of the investigations of textural motions and optical studies of the textures. It has been pointed out that the polygons are in many respects similar to the spherulites formed in the crystallization of various polymers.