CHAPTER I

INTRODUCTION

1.1 Nematic Liquid Crystals

Many organic compounds whose molecules are rod-like or disc-like, i.e., have shape anisotropy, exhibit mesomorphic or liquid crystalline phases (see for e.g., de Gennes, 1975; Chandrasekhar, 1977; de Jeu, 1980). They exhibit some properties of crystals and some others of liquids. All of our studies have been made on the simplest type of liquid crystal, viz., the nematic phase. The nematic liquid crystal (NLC) is characterised by an orientational order of the anisotropic molecules about a direction which is denoted by a unit vector $\hat{n}$ called the director (Fig.1.1). The centres of mass of the molecules have a liquid-like random distribution. The orientation of $\hat{n}$ can be controlled by external forces like those imposed by the cell walls. The director $\hat{n}$ is apolar with $+\hat{n}$ and -6 being indistinguishable. Therefore nematics are not ferroelectric. There is a two-fold rotational symmetry about any axis normal to $\hat{n}$ and reflection symmetry in planes parallel and perpendicular to $\hat{n}$. The nematic phase is optically uniaxial with the optic axis along 6.

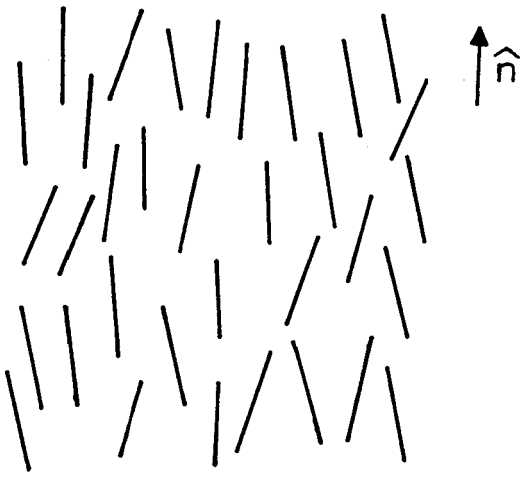

Fig.1.1: Schematic diagram showing the molecular arrangement in nematic liquid crystals.

1.2 The Orientational Order Parameter

In view of the apolar nature of the director, the orientational order parameter S is a second rank tensor. For cylindrically symmetric molecules it is defined as (Tsvetkov, 1942)

$$S = \frac{1}{2} < 3\cos^2\theta - 1 > \tag{1.1}$$

where θ is the angle between the molecular long axis of a rod-like molecule and the director $\hat{n}$. The brackets $< >$ indicate an ensemble average. From this expression, it is clear that for a perfect ordering S=1 and in the isotropic phase S=0. Usually the maximum value of S in a typical NLC ranges from 0.6 to 0.8. The value of S at the nematic-isotropic transition is usually about 0.3 to 0.5 and the nematic-isotropic phase transition has a first order character (de Gennes, 1975).

1.3 Curvature Elasticity

In nematic liquid crystals there is no translational order of the molecules. The director orientation of a nematic can gradually vary in space, with the magnitude of the order parameter remaining constant. Hence the fundamental elasticity of the NLC is associated with the curvature in the director field. The continuum theory of curvature elasticity was developed by Oseen (1933), Frank (1958) and Nehring and Saupe (1971). Taking into account the symmetry of the nematic phase there can be three elementary deformations, i.e., splay, twist and bend (Fig.1.2). The elastic free energy density, which is a scalar quantity can be expanded in terms of the director $\hat{n}$ and its gradients as,

$$f^d = \frac{1}{2}K_1(\nabla \cdot \hat{n})^2 + \frac{1}{2}K_2(\hat{n} \cdot \nabla \times \hat{n})^2 + \frac{1}{2}K_3(\hat{n} \times \nabla \times \hat{n})^2 \tag{1.2}$$

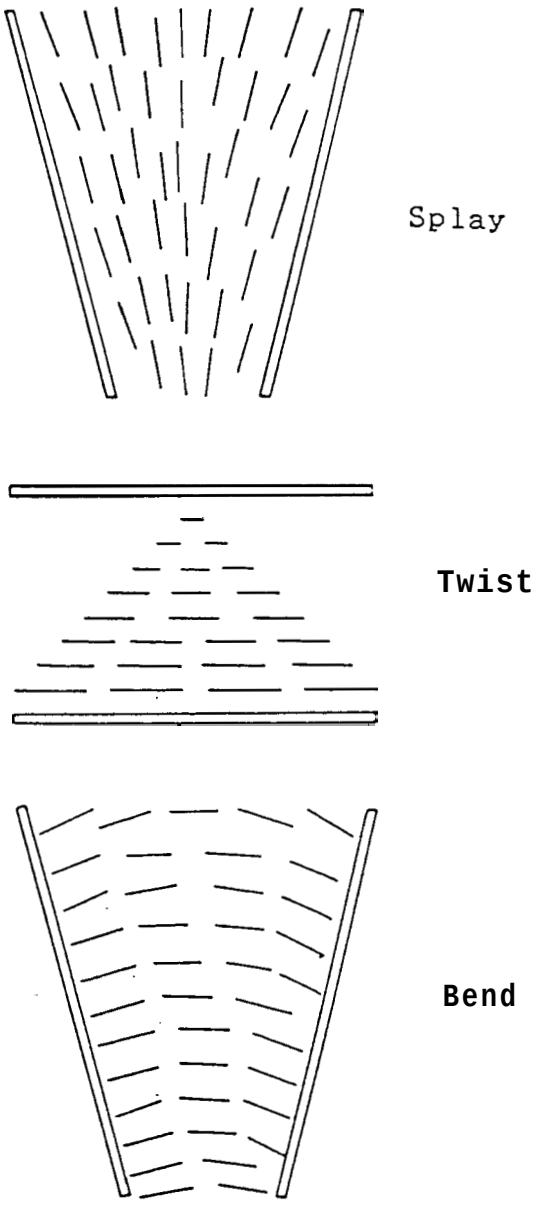

Fig.l.2: The three principal types of deformations in a nematic (after Chandrasekhar 1977).

where K_1, K_2 and K_3 are elastic constants corresponding to splay, twist and bend deformations respectively. Usually $K_3 > K_1 > K_2$. These constants are positive and are $\sim 10^{-11}$ newton (N). These elastic constants are highly temperature dependent and vary approximately as the square of the order parameter S.

In the absence of external fields, the condition for equilibrium can be obtained by minimising the energy given by equation (1.2) with respect to all variations of the director field $\hat{n}(\vec{r})$ and is given by (de Gennes, 1975)

$$h_i^d = -\left(\frac{\partial f^d}{\partial n_i}\right) + \partial_j \left(\frac{\partial f^d}{\partial g_{ji}}\right) = -C(\vec{r})n_i \tag{1.3}$$

where $\partial_j = \frac{\partial}{\partial x_j}$, $g_{ji} = \partial_j n_i$, $C(\vec{r})$ is an arbitrary function of $\vec{r}$ and $\vec{h}$ is called the molecular field. In equilibrium the director will be parallel to the molecular field at each point. Otherwise, when $\hat{n}$ and $\vec{h}$ are not parallel, the director experiences a torque which tends to make them collinear. The torque density is given by $\Gamma^d = A \times \vec{h}^d$. In the presence of an external field acting on the nematic, in addition to $\vec{h}^d$, the molecular field arising from the external field should also be considered.

Nematics have anisotropic physical properties. In the following we give a brief description of some of the physical properties of NLC, which will be relevant to our experiments.

1.4 Dielectric Anisotropy $\Delta\epsilon$

The dielectric displacement $\vec{D}$ induced in a nematic liquid crystal by an electric field $\vec{E}$ is given by (de Gennes, 1975)

$$\vec{D} = \epsilon_\perp \vec{E} + (\epsilon_\parallel - \epsilon_\perp)(\hat{n} \cdot \vec{E})\hat{n} \tag{1.4}$$

where $\epsilon_\parallel$ and $\epsilon_\perp$ are the principal dielectric constants parallel and perpendicular to the director respectively. The dielectric anisotropy $\Delta\epsilon (= \epsilon_\parallel - \epsilon_\perp)$ may be positive

or negative depending on the molecular structure. $\Delta\epsilon$ is positive for molecules with permanent dipole moments parallel to their long axes. It is negative ($\epsilon_{||} < \epsilon_{\perp}$) for molecules having permanent dipole moments at large angles to the long axes. The dielectric anisotropy is temperature as well as frequency dependent.

The free energy density of a nematic in an electric field is given by (de Gennes, 1975)

$$f^{\epsilon} = -\frac{\epsilon_{\perp} E^2}{8\pi} - \frac{\Delta\epsilon(\hat{n} \cdot \vec{E})^2}{8\pi} \tag{1.5}$$

The corresponding molecular field which is got by minimising f^{c},is given by

$$\vec{h}^{\epsilon} = \frac{\Delta\epsilon(\hat{n} \cdot \vec{E})\vec{E}}{4\pi} \tag{1.6}$$

Most of the nematic liquid crystals have diamagnetic anisotropy (AX), which is defined in a similar fashion. This property is useful to align NLCs using magnetic fields.

1.5 Electrical Conductivity

In nematic liquid crystals impurity ions give rise to a weak electrical conductivity of the medium. The anisotropy of conductivity A a $= \sigma_{||} - \sigma_{\perp}$, where $\sigma_{||}$ and $\sigma_{\perp}$ are the principal components of conductivity parallel and perpendicular to 6. Usually for nematics $\Delta\sigma$ is positive as the ions can move more freely along $\hat{n}$ than perpendicular to it. For an electric field E applied in an arbitrary direction, the current density is given by

$$\vec{J} = \sigma_{\perp}\vec{E} + \Delta\sigma(\hat{n} \cdot \vec{E})\hat{n}. \tag{1.7}$$

The electrical conductivity is a function of frequency of the applied AC field and also of the temperature of the nematic medium. Conductivity anisotropy plays

an important role in the formation of electrohydrodynamic instabilities in NLC (Chapters II and III).

1.6 Alignment of Liquid Crystals

In an unaligned nematic medium the director $\hat{n}$ varies gradually from point to point. However in experimental situations it is desirable to have a monodomain sample of a nematic, in which $\hat{n}$ is uniformly aligned. In a planar or homogeneously aligned sample, the director $\hat{n}$ is parallel to the bounding plates and hence the optic axis is parallel to the surfaces (Fig.1.3a). This alignment can be obtained by unidirectional rubbing of the glass plates (Chatelain, 1955; Guyon and Urbach, 1976) coated with a polyimide solution or by a vacuum coating of the plates with silicon monoxide at an oblique angle (Janning, 1972). The molecules sit in the grooves created by the rubbing action in polyimide coated plates, or in the grooves formed by the shadowing effect in SiO coated plates, to give rise to the planar alignment.

In the homeotropic alignment the director is aligned perpendicular to the bounding plates and hence the optic axis is normal to them (Fig.1.3b). This is obtained by treating the glass plates with certain polymers such as octadecyl triethoxy silane (ODSE), which have chemical groups that can attach themselves to the surface and have long chains which stick out of the surface.

Treating one of the plates for homogeneous alignment and the other for homeotropic alignment, we get a hybrid aligned nematic (HAN) cell. The director pattern in such a cell has a splay-bend distortion as shown in figure 1.3c. We make use of such a geometry in some of our experiments (Chapters IV and V).

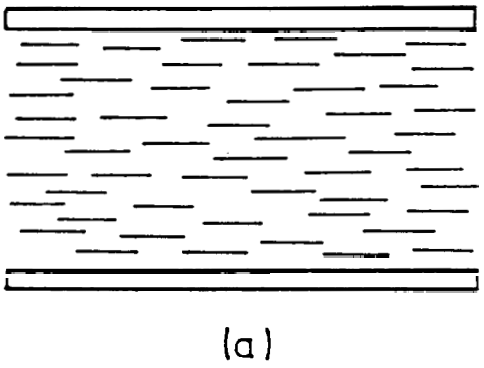

Fig.1.3: Orientation of the director ($\hat{n}$) in cells with (a) homogeneous alignment, (b) homeotropic alignment and (c) hybrid alignment.

1.7 Anchoring Energy

The anchoring energy measures the strength of anchoring of the director along a well defined direction, known as *easy axis* at the surface. It depends on the chemical nature of the NLC and that of the surface. The anisotropic part of the surface free energy density is written as (Rapini and Papoular, 1969)

$$F_s(\theta) = \frac{1}{2} W_\theta \sin^2(\theta - \theta_o)$$

where θ_o and 8 are the polar angles made by the easy axis (n_o) and the actual orientation of the director $(\hat{n})$ respectively with the surface normal. W_θ is the anchoring energy for tilt orientation of the director. In chapter V we will describe a method of estimating W_θ.

The anchoring energy (W_ϕ) for azimuthal orientation can also be defined in a similar way. **A** method of measuring W_ϕ will be described in Chapter VI.

In many experiments (see chapter III) external fields are used to induce distortions in surface-aligned monodomain samples. If the surface anchoring is weak, the orientation of $\hat{n}$ at the surface is affected by the bulk distortion. Then to determine the equilibrium configuration of the system the surface terms in the free energy will have to be considered. On the other hand, the surface terms can be neglected in the case of *strong* anchoring at the surface (see Chapters V and VI).

1.8 Hydrodynamics of Nematics

The hydrodynamic properties of NLCs are complex due to the orientational ordering of the molecules. This is because the translational motion of a molecule gets coupled to its rotational motion in the nematic phase. Flow can disturb the alignment of the director. Ericksen (1960, 1961, 1962) and Leslie (1966,1968) have developed a

continuum theory of the hydrodynamics of nematics. In an ideal liquid with zero viscosity (Landau and Lifshitz, 1953; Feynmann et al., 1964), the Euler equation is given by

$$\frac{\partial \vec{v}}{\partial t} + (\vec{v}\nabla)\vec{v} = -\frac{\nabla \vec{P}}{\rho} + \vec{g} + \frac{\vec{f}}{\rho} . \tag{1.8}$$

In this equation v is the velocity at a point (x,y,z), P is the pressure at this point. ρ the density of the liquid, g the acceleration due to gravity and f the external force. The equation of continuity is given by

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0 \tag{1.9}$$

The compressibility is quite small for liquids, and in the problems that we will discuss, they can be considered to be incompressible and hence $\frac{\partial \rho}{\partial t} = 0$ and ρ =constant. Hence equation (1.9) reduces to

$$\nabla \cdot \vec{v} = 0 \tag{1.10}$$

For a viscous liquid, an additional term to account for frictional force (f_{visc}) should be added to the Euler equation (1.8). Then

$$\frac{\partial \vec{v}}{\partial t} + (\vec{v}\nabla)\vec{v} = \left[-\frac{\nabla \vec{P}}{\rho} + \vec{g} + \frac{\vec{f}}{\rho}\right] + \frac{\vec{f}_{visc}}{\rho} \tag{1.11}$$

The viscous stress tensor (σ'_{ij}) is defined in terms of the gradients of velocity, as

$$\sigma'_{ij} = \eta \left(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i}\right) \tag{1.12}$$

where i, j = x,y,z and η is the viscosity of the liquid. By considering a cubic volume element of moving liquid and using the spatial derivatives of the stress tensor, the viscous force may be calculated as $\eta\nabla^2\vec{v}$. Then the resulting equation of motion for an isotropic liquid can be written as,

$$\frac{\partial \vec{v}}{\partial t} + (\vec{v}\nabla)\vec{v} = \left[-\frac{\nabla \vec{P}}{\rho} + \vec{g} + \frac{\vec{f}}{\rho}\right] + \eta\frac{\nabla^2\vec{v}}{\rho} . \tag{1.13}$$

This is the well known Navier-Stokes equation. This has been extended to NLCs by different groups and here we follow Leslie's approach (Leslie, 1966 and 1968).

Leslie took into account the dependence of shear stress not only on the velocity gradients but also on the orientation and the rate of rotation of the director. The viscous stress tensor in its most general form, obtained from symmetry arguments, is written as (de Gennes, **1975)**

$$\begin{aligned}\sigma'_{ij} &= \alpha_1 n_k n_l A_{kl} n_i n_j + \alpha_2 n_i N_j + \alpha_3 n_j N_i + \alpha_4 A_{ij} \\ &\quad + \alpha_5 n_i n_k A_{kj} + \alpha_6 n_j n_k A_{ki} \end{aligned} \tag{1.14}$$

where $\alpha_{(1-6)}$ are called the Leslie viscosity coefficients and they are of the order of 0.01 to 0.1 $\mathrm{Kgm^{-1}s^{-1}}$. $A_{ij} = \frac{1}{2}\left[\frac{\partial v_j}{\partial x_i} + \frac{\partial v_i}{\partial x_j}\right] = A_{ji}$ is the shear rate tensor.
The rate of motion of the director relative to the background liquid is given by

$$\vec{N} = \frac{\partial \hat{n}}{\partial t} - \vec{\omega} \times \hat{n} \tag{1.15}$$

where $\vec{\omega} = \frac{1}{2}(\mathrm{V} \times \vec{v})$ is an angular velocity.

$\vec{\Gamma}_{visc}$ is the torque per unit volume on the director due to viscous forces and can be written as (de Gennes, 1975)

$$\vec{\Gamma}_{visc} = -\hat{n} \times \vec{h}_{visc} \tag{1.16}$$

where $\vec{h}_{visc} = (\gamma_1 \vec{N} + \gamma_2 \bar{\bar{A}} \cdot \hat{n})$. $\gamma_1 = \alpha_3 - \alpha_2$ and $\gamma_2 = \alpha_6 - \alpha_5$. Parodi (1970) using Onsager's reciprocity relations showed that $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$. The α_4 term is independent of the director and corresponds to the viscosity (η) of an isotropic fluid. Now the equation of motion for the moving fluid can be written as

$$\rho\left(\frac{d\vec{v}}{dt}\right) = \vec{f} + (\vec{\nabla} \cdot \bar{\bar{\sigma}}) \tag{1.17}$$

The inertial term $\rho(d\vec{v}/dt)$ can be neglected in most problems. $\vec{f}$ is the external force acting on the fluid, for example due to the applied field $\vec{E}$ acting on a field-induced

space charge density Q (i.e., $\vec{f} = Q\vec{E}$). $(\nabla . \bar{\bar{\sigma}})$ is the force due to the total stress in the medium. Since small deformations are involved in the problems considered, we simplify equation (1.17) by retaining only the linear terms. Then we get

$$-\nabla\vec{P} + \nabla\cdot\bar{\bar{\sigma}}' + Q\vec{E} = 0 \tag{1.18}$$

The simplified coupled equations for tlie director and fluid velocity (Eqn.1.18) along with equation (1.10) are used in the study of EHD problems in later chapters of this thesis.

1.9 Flexoelectricity

A macroscopic polarisation can be induced in a nematic liquid crystal by splay arid bend distortions of the director field, as was first shown by Meyer (1969). This flexoelectric polarisation is given by

$$\vec{P} = e_1\hat{n}(\nabla\cdot\hat{n}) + e_3(\nabla\times\hat{n})\times\hat{n} \tag{1.19}$$

where e_1 and e_3 are the two flexoelectric coefficients corresponding to splay and bend deformations respectively. According to Meyer's model only nematics made up of polar molecules with shape assymmetry can be expected to exhibit flexoelectricity. For instance (Fig.l.4) a nematic consisting of pear shaped molecules with longitudinal dipole moments becomes polarised under a splay distortion and a nematic made of banana shaped molecules with transverse dipole moments becomes polarised under a bend distortion. However, in the undistorted state, because of equal probability of opposite orientations the dipole moments of tlie molecules cancel one another and the net dipole density is zero. For weak distortions the flexoelectric polarisation is given by tlie equation (1.19). This expression is the most general polar vector that

can be constructed from the apolar director field $\hat{n}$, arid its first order derivatives. e_1 and e_3 have the dimensions of charge/length.

Another microscopic model was presented by Prost and Marcerou (1977) in which the contributions to flexoelectric effect from quadrupole moments of the molecules are considered. In the undistorted state (Fig.1.5a), there is no net dipole moment due to the symmetry of the arrangement of the quadrupoles. But when there is splay deformation, there is a net dipole moment (Fig.1.5b) and hence the medium is polarised. Though this quadrupolar contribution to flexoelectric effect is of the same magnitude as due to dipolar contribution, the former is independent of the shape of the molecules. Since all nematogenic molecules have finite quadrupole moments, flexoelectric effect is an universal property of nematics.

In the presence of an external electric field E, the polarisation P couples linearly with E leading to a free energy density

$$f^{fl} = -\vec{P} \cdot \vec{E} \tag{1.20}$$

This is minimiscd to get the corresponding molecular field

$$\vec{h}^{fl} = e_1\{\vec{E}(div\hat{n}) - grad(\vec{E} \cdot h)\} + e_3\{\vec{E} \times (curl\, h) - curl(\vec{E} \times h)\} \tag{1.21}$$

The linear dependence of the flexoelectric energy on the gradient in the director field has some interesting consequences. For instance if the distortiori is planar represented by the angle $\theta(z)$, then f^{fl} identically satisfies the Euler-Lagrange equation

$$\left(\frac{\partial f^{fl}}{\partial \theta}\right) - \partial_z \left(\frac{\partial f^{fl}}{\partial \theta_{,z}}\right) = 0 \tag{1.22}$$

where $\partial_z = \left(\frac{\partial}{\partial z}\right)$ and $\theta_{,z} = \left(\frac{\partial \theta}{\partial z}\right)$, and therefore does not give rise to any volume torque. When there is weak anchoring at the surface the flexoelectric effect results in a surface torque. This effect is used in the estimation of tlie anchoring strengths

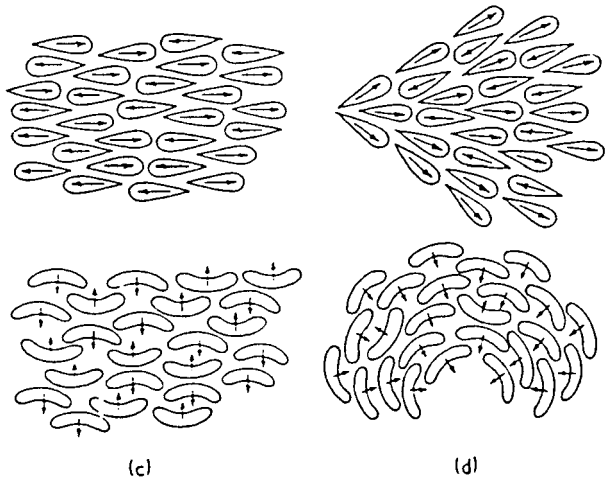

Fig.1.4: Meyer's model of flexoelectricity. The nematic cornposed of asymmetric polar molecules has no net polarization in the undeformed state (a and *c),* but develops a polarization under splay (b) and bend (d) deformation. (after Mcycr 1969).

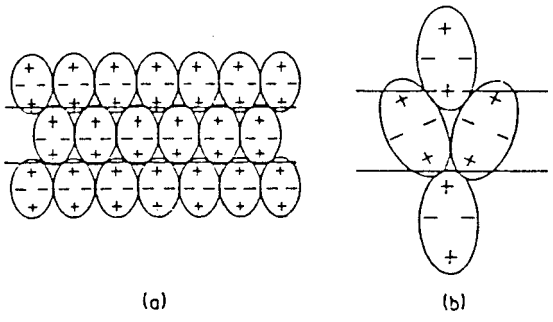

Fig.1.5: (a) A regular stacking of quadrupoles. Because of the symmetry of the arrangement of the quadrupoles there is no bulk polarisation. (b) The structure becomes polarised when subjected to a splay distortion. (after Prost and Marcerou, 1977).

in a HAN cell (Chapter V). However if the distortion is non-planar, and is described by the angles $\theta(z)$ and $\phi(z)$, then f^{fl} gives rise to a bulk torque. This is made use of in the determination of flexoelectric coefficients of NLC (Chapter IV).

1.10 Electrohydrodynamic Instabilities in Nematics

Convective hydrodynamic instabilities are well known in isotropic liquids. For example, in Rayleigh-Benard instability (see for example Dubois-Violette et ***al.,*** 1978) a regular convective flow of liquids sets in when an adverse temperature gradient exceeds a certain value. In this case the convection sets in due to the action of the gravitational field on a vertical density gradient.

Hydrodynamic instabilities are also observed in weakly conducting isotropic liquids under the action of DC or AC fields (Blinov, 1983). Felici (1969) proposed a mechanism in which unipolar charge injection by one of the electrodes results in a lion-uniform distribution of space charges (Fig.1.6). The interactiori of the external field with these charges gives rise to a force which destabilises tlie medium resulting in a cellular flow in the medium. However this instability disappears above a frequency τ^{-1}, where τ is the transit time for injected charge carriers.

Convective instabilities are all the more interesting in tlie case of nematic liquid crystals with negative or slightly positive $\Delta\epsilon$ and positive $\Delta\sigma$ (see for e.g., de Gennes, 1975). Under tlie action of an external electric field such NLC exhibit convective instabilities, called electrohydrodyamic (EHD) instabilities, which were first observed during 1930s (Freedericksz and Zolina, 1933 and Freedericksz and Tsvetkov, 1934). But detailed studies were made only in the late 60s by which time the possibility of using EIID instabilities in display devices was recognised. A typical experimental set up (Fig.l.7) to study EHD instabilities in NLCs consists of two transparent

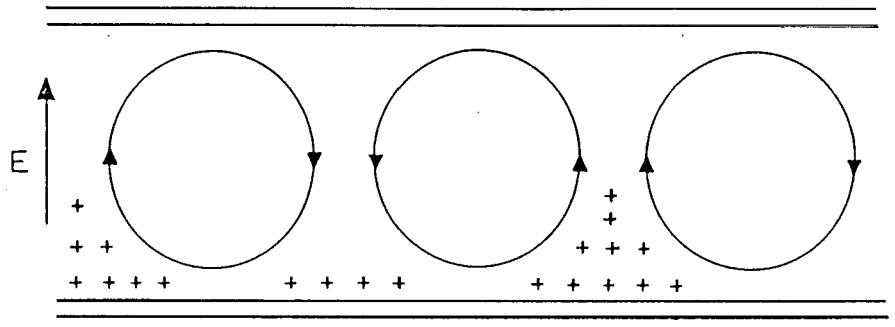

Fig.l.6: Schematic diagram showing unipolar charge injection from one of the electrodes illustrating Felici mechanism.

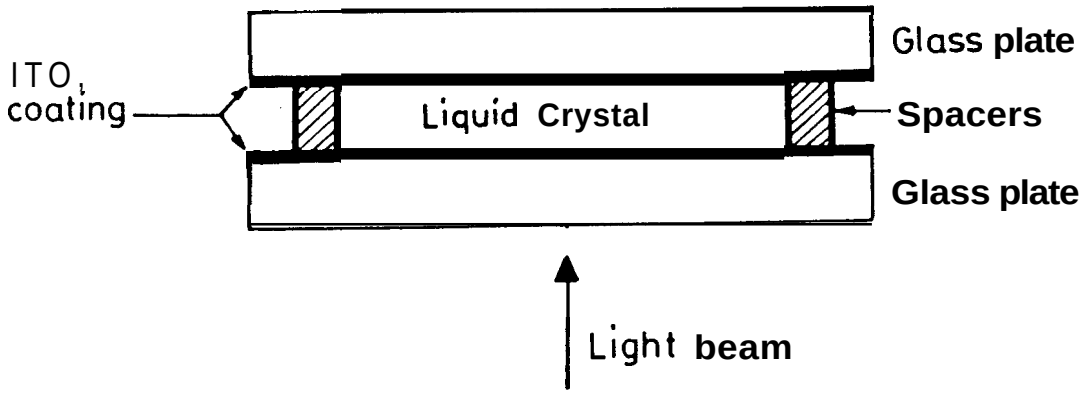

Fig.l.7: Experimental arrangement for observing Williams Domain Mode (WDM).

conducting glass plates, which are treated to get a homogeneous alignment of the nematic director along the X-axis, say. These are used in the construction of a cell of thickness $\sim 20\mu m$. The cell is filled with an appropriate NLC. Now with the application of a voltage above a threshold value between the glass plates, i.e., along the Z axis, the medium gives rise to an optical pattern (Fig.1.8) consisting of regular, parallel striations, perpendicular to the undistorted director, i.e., along the Y axis. This is called the Williams Domain Mode (WDM) (Williams, 1963). The important observations on this instability may be summarised as follows:

a. WDM is characterised by a threshold voltage which is independent of cell thickness.

b. WDM is observable under DC fields or low frequency AC fields.

c. The width of these domains is of the order of the cell thickness.

d. There is cellular fluid motion within the domains which is clearly seen by the movement of tracer dust particles. Further, fluid motion has opposite vorticity in adjacent domains (Fig.1.9).

1.10.1 The Carr-Helfrich Mechanism

Carr (1963) suggested that anisotropy of conductivity may give rise to space charge formation due to ion segregation under the action of an external electric field in the liquid crystal. This was made use of by Helfrich (1969) while proposing a mechanism for Williams domain instability.

Consider an infinitesimal bend deformation in a homogeneously aligned nematic film with negative dielectric anisotropy **(Ac)** arid positive conductivity anisotropy

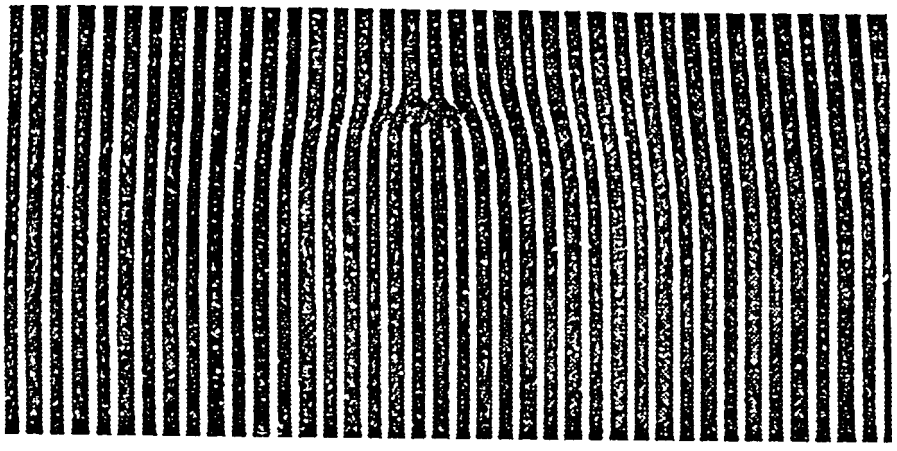

Fig.l.8: Williams Domain Mode (WDM) at a voltage slightly above the threshold value. $\hat{n}$ indicates the direction of alignment of the director (d = $40\mu m$). (Ref. Blinov, 1983).

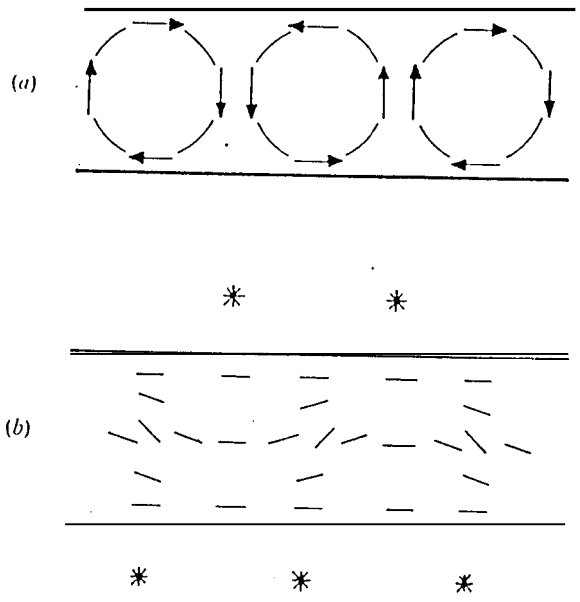

Fig.1.9: Schematic diagram of Williams domains indicating (a) flow and (b) tlirector orientation. ⋆ ⟶ indicate bright foci.

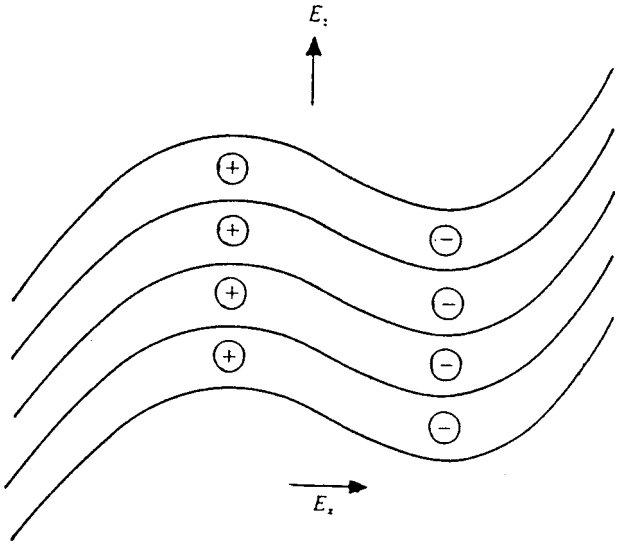

Fig.1.10: Space charge formation in an applied field E_z caused by a bend fluctuation of the director in a NLC with positive Aa. E_x is the resulting transverse field.

(Au). If it is subjected to a DC electric field (E) between the glass plates, i.e., along the Z-axis (Fig.1.10), the ions move more easily along the director than in a perpendicular direction, since $\sigma_{\parallel} > \sigma_{\perp}$. This results in space charge formation giving rise to an internal transverse electric field (E_x) along the X axis. Tlie external field E acting on these space charges (Q) produces a force, which tends to set them in motion resulting in a bulk flow of the medium, when the field E is sufficiently large. This hydrodynamic torque along with the dielectric torque due to internal electric field E_x produces a destabilising effect on the orientation of the dircctor, while the dielectric torque due to the external field E and the elastic torque have stabilising effects on the director orientation. Hence if E is sufficiently large, an infinitesimal bend distortion in the director field grows and the system becomes unstable. This results in a breaking up of the medium into a number of hydrodynamic rolls or domains. This cellular flow of the medium produces a static periodic deformation of the director field and gives rise to the optical pattern as described before.

A linear one dimensional theory of EHD instabilities in which the boundary conditions are ignored was worked out by Helfrich (1969). The medium is assumed to be of an infinite extent in the XY plane so that the lateral boundary conditions may be neglected. An infinitesimal bend fluctuation of tlie director field is considered in the XZ plane with the director $\hat{n}$ making a small angle θ with the X-axis. Let Q be tlie space charge density created in tlie medium. As long as tlie boundary conditions are neglected, **0** and all other quantities depend only on X. Because of this simplification, only the Z-component of velocity appears in the equations. Retaining only the linear terms, the director in the distorted state is given by

$$\hat{n} = (\cos\theta, 0, \sin 0) \simeq (1, 0, \theta)$$

The system is described by the following equations:

1. The Poisson equation $div\,\vec{D} = 4\pi Q$, where $\vec{D}$, the displacement vector is given by

$$\vec{D} = \epsilon_\perp \vec{E} + (\epsilon_\parallel - \epsilon_\perp)(\hat{n} \cdot \vec{E})\,\hat{n}\ ,$$

substituting for $\vec{D}$ in the Poisson equation and simplifying, we get

$$\epsilon_\parallel \left(\frac{dE_x}{dx}\right) + \Delta\epsilon E \left(\frac{d\theta}{dx}\right) = 4\pi Q \tag{1.23}$$

2. As we are considering a DC applied field the charge conservation equation is given by

$$div(\vec{J}) = 0$$

where $\vec{J}$ is the current density given by $\vec{J} = \sigma_\perp \vec{E} + \Delta\sigma(\hat{n} \cdot \vec{E})\hat{n}$. From these two equations we get

$$\sigma_\parallel \left(\frac{dE_x}{dx}\right) + \Delta\sigma E \left(\frac{d\theta}{dx}\right) = 0 \tag{1.24}$$

3. Using equation (1.18) the equation of motion in the linear regime is given by

$$-\eta \left(\frac{d^2 v_z}{dx^2}\right) = QE \tag{1.25}$$

where $\eta = \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2)$ and a's are viscosity coefficients.

4. The torque balance equation is given by

$$\vec{\Gamma}^d + \vec{\Gamma}^\epsilon = \vec{\Gamma}^{hy}$$

where $\vec{\Gamma}^{hy}$ is the hydrodynamic torque, $\vec{\Gamma}^d$ the elastic torque and $\vec{\Gamma}^\epsilon$ the dielectric torque. This reduces to

$$K_3 \left(\frac{d^2\theta}{dx^2}\right) + \left(\frac{\Delta\epsilon\sigma_\perp}{4\pi\sigma_\parallel}\right) E^2\theta - \alpha_2 \left(\frac{dv_z}{dx}\right) = 0 \tag{1.26}$$

Eliminating E_x, v_z and Q in terms of θ from equations (1.23) to (1.26) we get

$$K_3\left(\frac{d^2\theta}{dx^2}\right) - \left(\frac{\alpha_2\epsilon_{\|}}{4\pi\eta}\right)\left(\frac{\Delta\sigma}{\sigma_{\|}} - \frac{\Delta\epsilon}{\epsilon_{\|}}\right)E^2\theta + \left(\frac{\Delta\epsilon\sigma_{\perp}}{4\pi\sigma_{\|}}\right)E^2\theta = 0 \qquad (1.27)$$

In this model, the rolls are assumed to be formed with their wavevector along $\hat{n}$. Taking the solution of the form $\theta = \theta_o \cos(qX)$, we get for the threshold field

$$E_{th}^2 = \frac{4\pi q^2 K_3}{\left[\Delta\epsilon\frac{\sigma_{\perp}}{\sigma_{\|}} - \frac{\alpha_2\epsilon_{\|}}{\eta}\left(\frac{\Delta\sigma}{\sigma_{\|}} - \frac{\Delta\epsilon}{\epsilon_{\|}}\right)\right]} \qquad (1.28)$$

Thus the threshold field is a function of the wavevector (q) of the distortion. The value of q cannot be predicted correctly by this model since the boundary conditions are not taken into account. Since the distortion energy is larger for larger q, while the dissipation due to the transverse flow is larger for smaller q, physically we can assume that the width of the domains must be comparable to the thickness of the nematic layer d. Hence putting $q = (\pi/d)$ in equation (1.28), a voltage threshold is obtained:

$$V_{th}^2 = \frac{4\pi^3 K_3}{\left[\Delta\epsilon\frac{\sigma_{\perp}}{\sigma_{\|}} - \frac{\alpha_2\epsilon_{\|}}{\eta}\left(\frac{\Delta\sigma}{\sigma_{\|}} - \frac{\Delta\epsilon}{\epsilon_{\|}}\right)\right]} \qquad (1.29)$$

This expression is found to be in qualitative agreement with experiments (Blinov, 1983). An extension of Helfrich model including tlie boundary conditions was developed by Penz and Ford (1972). The results broadly support the Helfrich model.

1.10.2 Excitation with AC Electric Fields

Depending on the frequency of the applied AC field the GIID instabilities can be classified into (a) conduction and (b) dielectric regimes.

The conduction regime is observed at frequencies below a certain cut off frequency (f_c) (Fig.1.11). The pattern observed in this regime is similar to WDM having a voltage threshold.

The dielectric regime or fast turn off mode is observed at frequencies above the cut off frequency (f_c) (Heilmeier and Helfrich, 1970 and Orsay Liquid Crystal Group, 1970, 1971). Unlike in the case of WDM, here the domain width is considerably less than the thickness of the nematic film. On increasing the applied voltage above the threshold value, these domains bend and niove giving rise to what arc called *Chevron Patterns* (Fig.1.12). This regime has a field threshold E_{th} in contrast to the voltage threshold V_{th} observed in the conduction regime. The threshold field varies with frequency of the applied field. Further the cut-off frequency f_c increases with the conductivity of the sample.

Dubois-Violette *et al.* (1971) and Smith *et al.* (1975) extended the Carr-Helfrich model to explain EHD instabilities under AC field excitation. Two coupled linearised equations for the space charge density Q and curvature $\psi = \dfrac{\partial \theta}{\partial x}$ are obtained:

$$\dot{Q} + \left(\frac{1}{\tau}\right) Q + \sigma_H E \psi = 0 \tag{1.30}$$

and

$$\dot{\psi} + \left(\frac{1}{T}\right) \psi + \frac{E}{\eta} Q = 0 \tag{1.31}$$

where

$$\frac{1}{\tau} = 4\pi \left(\frac{\sigma_{\|}}{\epsilon_{\|}}\right) \text{ is the relaxation rate of the space charges}$$

$$\frac{1}{T} = \frac{\eta_1}{\gamma_1 \eta_2} \left[- \frac{\epsilon_{\perp} \Delta\epsilon E^2}{4\pi \epsilon_{\|}} \right] \text{ is the relaxation rate of the director}$$

$$\frac{1}{\eta} = - \frac{\eta_1}{\gamma_1 \eta_2} \left[\frac{\Delta\epsilon}{\epsilon_{\|}} + \frac{\alpha_2}{\eta_1} \right] ; \quad \sigma_H = \sigma_{\|} \left(\frac{\epsilon_{\perp}}{\epsilon_{\|}} - \frac{\sigma_{\perp}}{\sigma_{\|}} \right)$$

$$\eta_1 = \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2) \text{ , arid } \quad \eta_2 = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6) - \frac{\alpha_3^2}{\gamma_1}$$

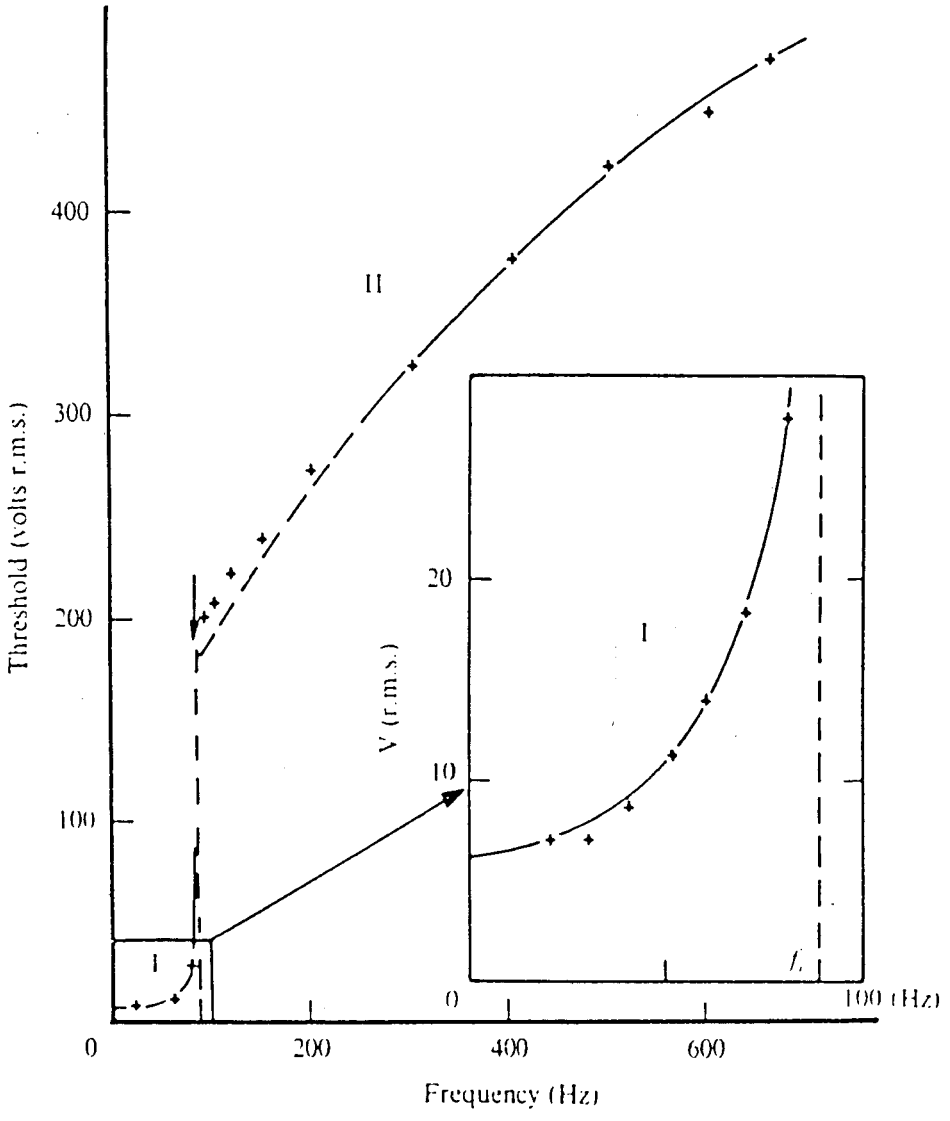

Fig.1.11: The threshold voltage of the AC instabilities in MBBA as a function of the frequency of the applied field. Regions I and II correspond to the conduction regime and the dielectric regime respectively. (Orsay Liquid Crystal Group, 1970).

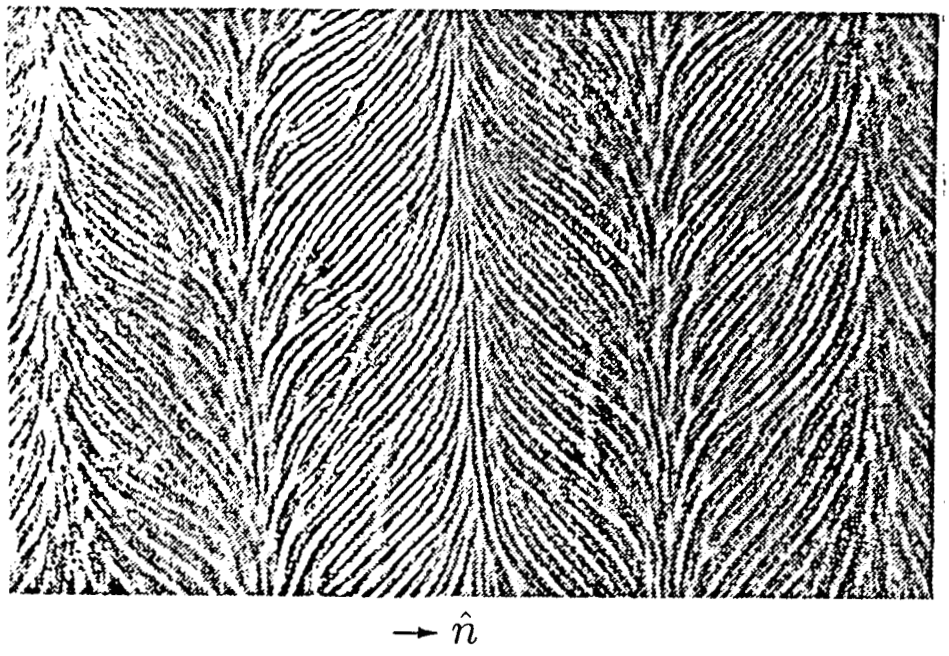

Fig.1.12: Chevron pattern observed at a voltage slightly above the threshold value in the dielectric regime. (from Blinov 1983).

Here $\gamma_1 = (\alpha_3 - \alpha_2)$, and the α's are Leslie viscosity coefficients.

From equations (1.30) and (1.31) it is clear that as E changes sign after every half a cycle, either Q or ψ should reverse its sign. In the first instance Q oscillates with the field, while ψ does not oscillate, i.e.,

$$Q\left(t + \frac{1}{2f}\right) = -Q(t) \text{ and } \psi\left(t + \frac{1}{2f}\right) = \psi(t)$$

This regime is called the conduction regime. On the other hand if ψ oscillates with the field while Q docs not, i.e.,

$$\psi\left(t + \frac{1}{2f}\right) = -\psi(t) \text{ and } Q\left(t + \frac{1}{2f}\right) = Q(t)$$

we get the dielectric regime.

A simplified one dimensional linear analysis was developed by Srriitli et *al.* (1975) for square wave excitation. Figure 1.13 gives the variation of threshold field for EHD instability with tlie frequency of the applied field. The essential requirement for the occurrence of the EHD instabilities in the conduction regime is that the director relaxation time (T) must be longer than the charge relaxation time (τ) (i.e., $T > \tau$) so that tlie space charges can develop in the medium. Further in this regime $\tau < \frac{1}{w}$ where w is the frequency of the applied field, and hence charges follow the field E (curve a). This is similar to the DC field case. Since $\frac{1}{T} = -\frac{\eta_1}{\gamma_1 \eta_2}\left(\ldots - \frac{\epsilon_\perp \Delta\epsilon E^2}{4\pi\epsilon_{||}}\right)$ and $\Delta\epsilon$ is negative, an increase in E results in a decrease of T. When T becomes comparable to τ we get a restabilisation branch (curve b). At frequencies higher than $1/\tau$, the charges cannot follow the field. In this case the curvature of the director can follow the field E if T is sufficiently small. This can be realised by increasing both the wave vector q and the field E. For T less than $1/\omega$ and hence also less than τ, tlie dielectric instability can set-in. Bodenscliatz' et ***al.*** (1988) have made detailed calculatioris for the AC case taking into accourit rigid boundary conditions.

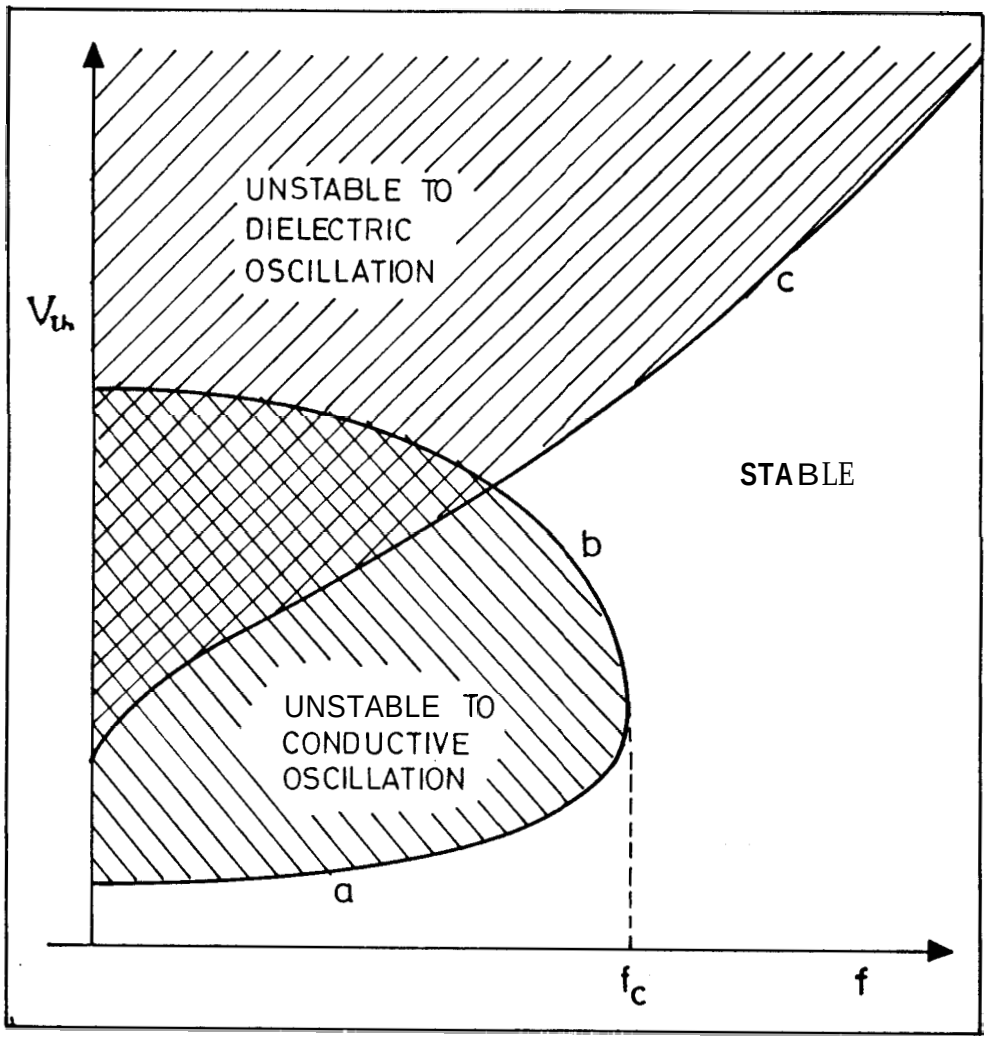

Fig.1.13: The variation of the threshold field with the frequency of the applied electric field, obtained from numerical calculations for square wave excitation. The curves labelled a,b and c correspond to the onset of the conduction regime, the restabilization branch and the onset of the dielectric regime, respectively. (from Smith et *al.* 1975).

In Chapter III we discuss EHD instabilities in a different geometry. Here the field is applied in the plane of a homogeneously aligned NLC film, such that it acts in a direction perpendicular to the director. We discuss an extension of the theoretical model to this geometry and also report some experimental results.

1.10.3 Oblique-Roll Instability

It is observed in experiments under a low frequency AC or DC excitation that EHD rolls are not normal to the undistorted director $\hat{n}$, but make an angle α with $\hat{n}$ (Hilsum and Saunders, 1980; Hirata and Tako, 1981 and Ribotta *et al.*, 1986). These are referred to as oblique rolls and the angle α is the angle between the wavevector of the oblique rolls and $\hat{n}$ (Fig.1.14). The following observations have been made on the oblique rolls:

1. The oblique rolls are formed right at the threshold of instability at low frequencies, up to a certain critical frequency f_o.

2. The obliquity of the rolls decreases as the ternperature increases. After a certain temperature they become normal rolls.

3. f_o increases with the conductivity of the sample.

4. Beyond f_o normal rolls are obtained at the threshold in the conduction regime. But as the field is further increased the normal rolls first become undulatory and then oblique.

The oblique rolls are now understood as arising from the flexoelectric effect (Madhusudana et al., 1987).

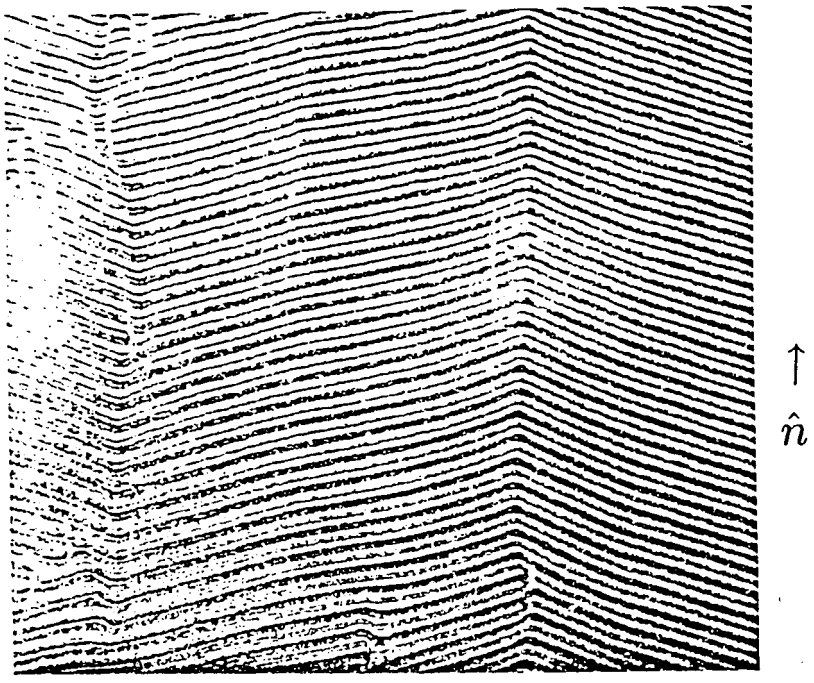

Fig.1.14: A photograph of oblique rolls. (from Hilsum and Saunders, 1980).

1.10.4 Travelling Wave Instability

A travelling wave instability has been observed in the conduction regime, especially in thin samples (Rehberg et al., 1988). In this type of instability, the EHD pattern moves parallel to the wavevector with equal probability of motion in either of the two possible directions. However the theoretical models developed until now do not give solutions corresponding to this type of instability. We have found that by changing the symmetry of the nematic cell by introducing a small pretilt angle at the bounding surfaces a propagating EHD instability under a DC field can be obtained. The direction of propagation depends on the direction of the applied field. This type of propagating EHD mode is due to additional flexoelectric torques which are $\pi/2$ radians out of phase with the main torques responsible for EHD instabilities. We describe this mode in Chapter II.

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