

Chapter 1

Introduction

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1.1 LIQUID CRYSTALS

Liquid crystals are states of matter whose symmetry and mechanical properties are intermediate between those of crystalline solids and isotropic liquids [1-3]. Hence they are also known as mesophases. They can be broadly classified into two classes: thermotropic liquid crystals and lyotropic liquid crystals. In thermotropic liquid crystals the transitions to the mesophases take place due to purely thermal effects. Lyotropic liquid crystals, on the other hand, occur in solutions for certain range of concentration of the solute molecules. Geometrical shape anisotropy of the constituent molecules is an essential criterion for any material to exhibit mesomorphism. For example, in the case of thermotropic liquid crystals they are either rod-like or disc-like [4].

1.2 CLASSIFICATION OF THERMOTROPIC LIQUID CRYSTALS

Based on their symmetry, thermotropic mesophases of rod-like molecules are classified into two classes: nematic and smectic.

The nematic mesophase is characterized by orientational ordering of the molecules and liquid-like

arrangement of their centres of mass (Fig.1a). The long axes of the molecules are oriented, on an average, parallel to a well-defined direction in space. This mean alignment direction is denoted by a unit vector \hat{n} , called the director! The direction of \hat{n} in space is arbitrary, and is imposed by minor forces such as the guiding effect of the walls of the container. The system is generally uniaxial, with infinite rotational symmetry about \hat{n} . There is also reflection symmetry in planes parallel and normal to \hat{n} , and two fold rotational symmetry about any axis normal to \hat{n} . This implies that \hat{n} and $-\hat{n}$ are physically equivalent. Consequently, nematics are not ferroelectric, even if the individual molecules carry a permanent electric dipole moment parallel to their long axis.

If the molecules are optically active, one obtains a modification of the nematic phase known as cholesteric or chiral nematic. This phase can also be obtained by doping a nematic with small amounts of an optically active compound. The lack of reflection symmetry at the molecular level results in a macroscopic chiral structure, viz, a helicoidal arrangement of the molecules (Fig.1b). This structure can be described by the director field, $n_x = \cos(q_0 z)$, $n_y = \sin(q_0 z)$ and $n_z = 0$.

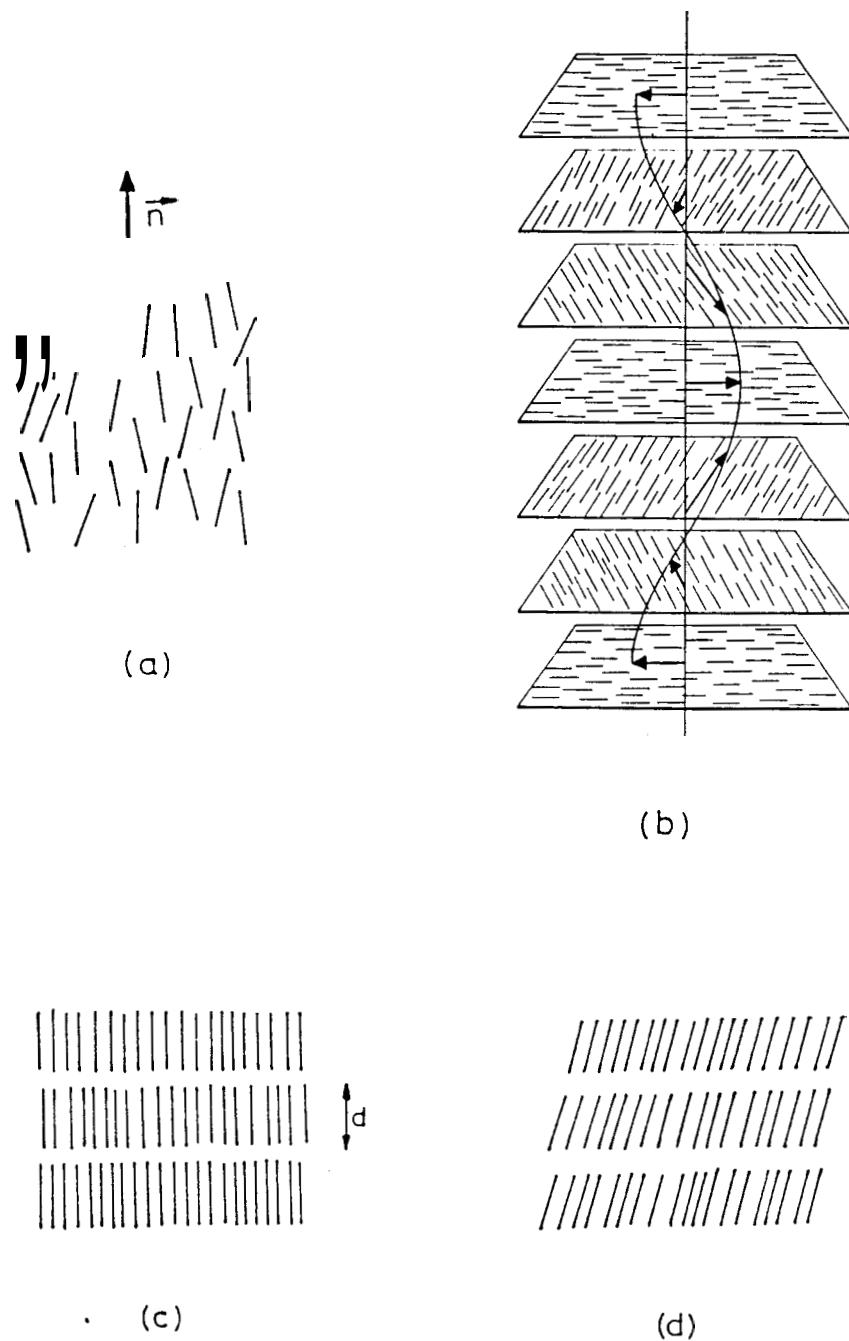


Fig.1. Schematic diagram showing the molecular arrangement in different types of mesophases exhibited by rod-like molecules. (a) nematic, (b) cholesteric, (c) smectic A and (d) smectic C.

The torsion of the helix, q_0 is in practice small, so that the pitch of the helix $P=2\pi/q_0$ is generally of the order of a few microns. The system is optically uniaxial with the optic axis along the axis of the helix.

The smectic phases are characterized by one degree of translational order with the molecules arranged in layers. As shown by Landau and Peierls [5], the thermal fluctuations in such a system can destroy the long range order. However, as pointed out by de Gennes [6], the smectic structure is readily stabilized by an external magnetic field or by the boundaries. Depending on the arrangement of the molecules within the layers, they are further classified as smectic A,B,C etc.

In the smectic A phase the molecules are arranged in layers with \hat{n} normal to the layers. Within each layer the centres of mass of the molecules have a two-dimensional liquid-like arrangement (Fig.1c). The system is uniaxial with infinite rotational symmetry about \hat{n} . Further, \hat{n} and $-\hat{n}$ are physically equivalent. Mesogenic compounds with chiral molecules can also exhibit the smectic A phase. In such cases the infinite rotational symmetry about \hat{n} does not imply a plane of symmetry parallel to \hat{n} .

The smectic C phase is similar to smectic A except that the molecules are tilted with respect to the layer normal (Fig.1d). The tilt is described in terms of a unit vector \hat{c} which is the projection of \hat{n} on the plane of the layer. It is usually called the c-director. The c-directors of adjacent layers are parallel on an average, ie, the c-director has long range order. An aligned sample of smectic C is optically biaxial. The symmetry elements of a smectic C are a twofold axis (normal to the $\hat{n}\hat{c}$ plane) and a plane of symmetry normal to it. As in a nematic, molecular chirality induces torsion in a smectic C. The c-director traces out a helix in space, with the axis of the helix normal to the layers. This is known as the chiral smectic C phase [7].

Thermotropic mesophases made up of disc-like molecules are of three types: nematic, columnar and lamellar [8,9] (Fig.2). A discotic nematic is also characterized by orientational ordering of the symmetry axes of the molecules, with the discs aligned more or less parallel to each other. If the molecules are chiral we get the chiral nematic phase with a helical arrangement of the director, as in the case of rod-like molecules. In the columnar mesophases the molecules are arranged in columns and depending on the symmetry of the packing of the columns

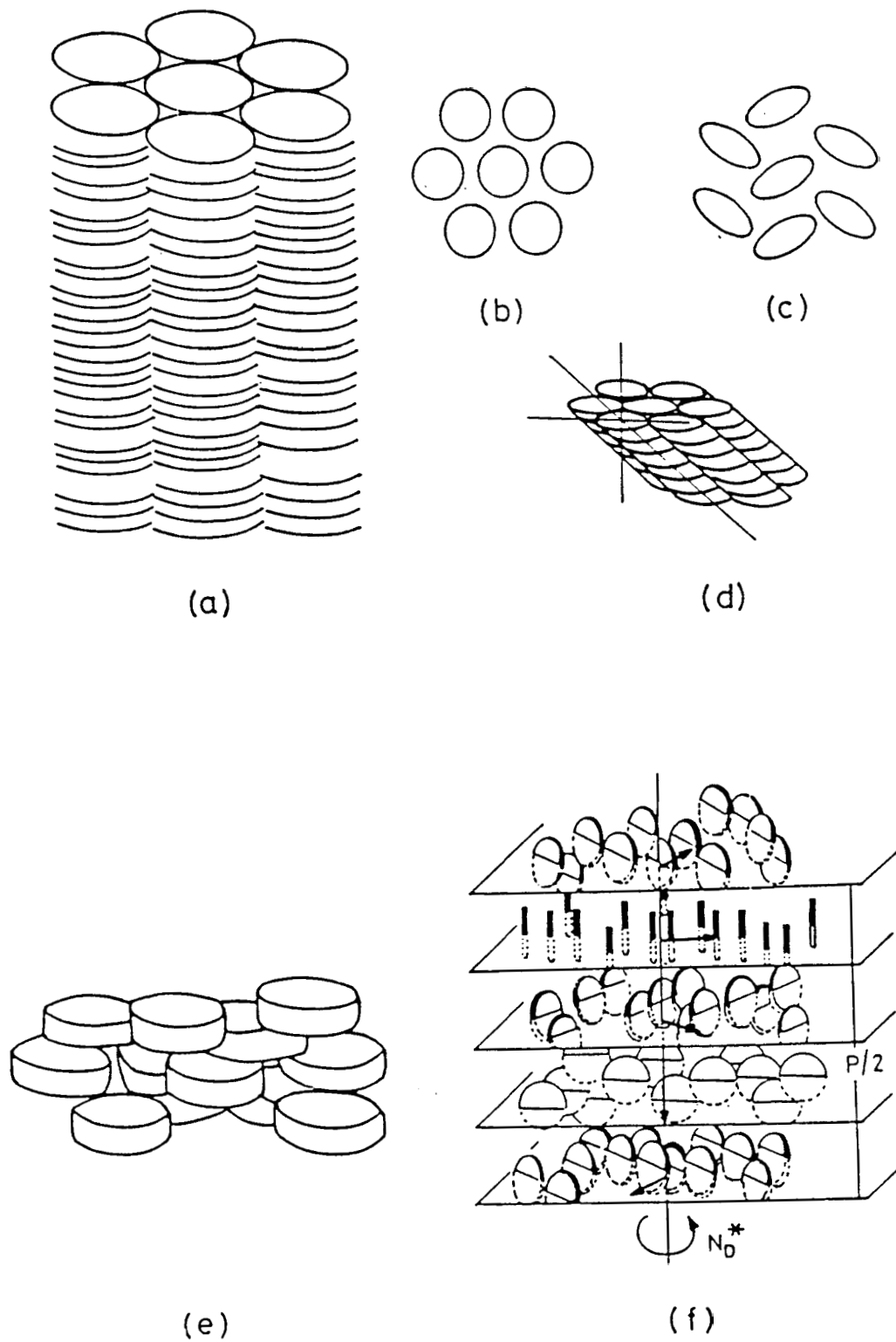


Fig.2. Schematic diagram showing the molecular arrangement in some discotic mesophases. (a) columnar phase, (b) hexagonal and (c) rectangular modifications of the columnar structure, (d) tilted columnar phase, (e) nematic and (f) chiral nematic.

they are classified as hexagonal or rectangular columnar phases. In addition, there is also a tilted columnar phase, where the axis of the columns is tilted with respect to the normal to plane containing the molecules. In the lamellar phase the molecules are arranged in layers, but the detailed structure has not yet been resolved.

1.3 THE NEMATIC PHASE

The nematic mesophase is characterized by orientational ordering of the molecules. As mentioned earlier this ordering can be described in terms of a unit vector \hat{n} , called the director, which gives the average alignment direction. In the case of nematics of rod-like molecules, \hat{n} gives the average orientation of the long axis of the molecule and in the case of discotic nematics it gives the average orientation of the short axis normal to the disc. The direction of \hat{n} can vary gradually from point to point in the medium. The system is uniaxial with the optic axis along \hat{n} . All the studies reported in the main body of this thesis are on the nematic phase. Further, the electrohydrodynamic instabilities in nematics are strongly influenced by many of its physical properties. Hence we briefly discuss the physical properties of nematics below.

The Orientational Order Parameter

The ordering in a nematic phase, assumed to be made up of cylindrically symmetric molecules can be described by the distribution function $f(\theta)$ for the angle between the director and the unique axis of the molecule [10,11]. $f(\theta)$ can be expanded in Legendre polynomials as:

$$f(\theta) = \frac{1}{2} + \sum_{n=1}^{\infty} \frac{1}{2} (4n+1) \langle P_{2n} \rangle P_{2n}(\cos\theta)$$

$$\text{where } \langle P_{2n} \rangle = \int_{-1}^1 P_{2n}(\cos\theta) f(\theta) d(\cos\theta).$$

Since \hat{n} and $-\hat{n}$ are equivalent, only the even powers of $\cos\theta$ appear in the above expansion. For an isotropic liquid all $\langle P_{2n} \rangle = 0$, and for perfect alignment of the molecules $\langle P_{2n} \rangle = 1$. Experimentally, the leading order parameter $\langle P_{2n} \rangle = \langle \frac{1}{2}(3 \cos^2\theta - 1) \rangle$ is easily measured and is normally referred to as the order parameter S of the nematic. The experimentally observed variation of S with temperature is shown in Fig.3 [12]. The value of S at the nematic-isotropic transition is usually about 0.3 to 0.5, indicating that the transition is first order [5].

Curvature Elasticity

In a nematic, the director orientation can vary gradually in space, with the magnitude of the order

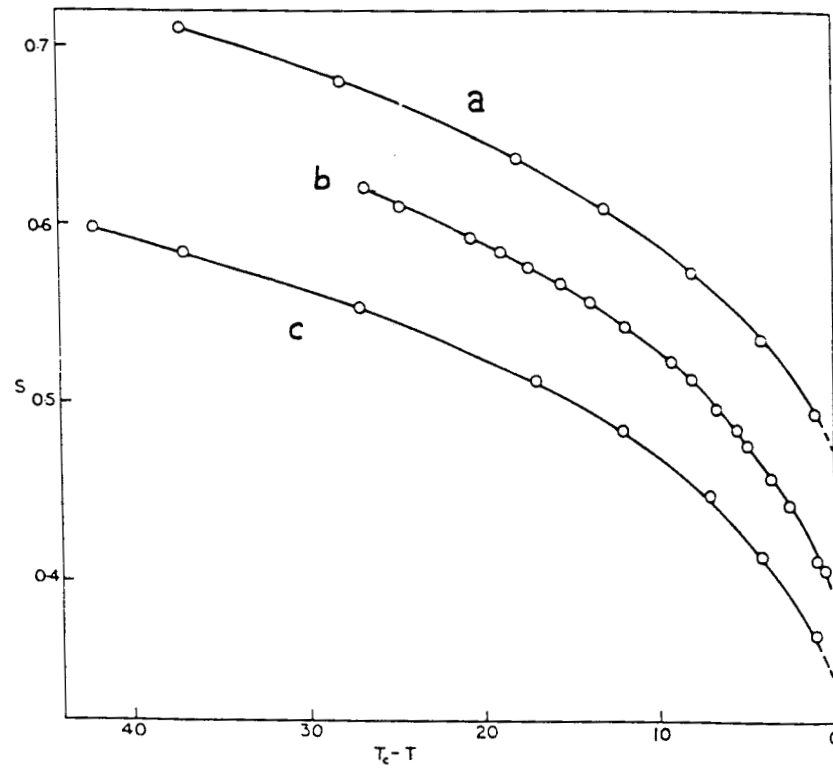


Fig.3. Variation of the orientational order parameter with temperature in p-azoxyphenetole (a), anisaldazine (b) and p-azoxyanisole (c). T_c is the nematic-isotropic transition temperature. (from Ref.12).

parameter remaining constant throughout the sample. The structure can be described by specifying the director at each point in the medium. Since it is a fluid, the fundamental elasticity in a nematic is associated with curvature in the director field, rather than with molecular displacements. A continuum theory of the curvature elasticity has been developed by Oseen [13], Frank [14] and Nehring and Saupe [15].

To describe the state of curvature at any point, we perform a Taylor series expansion of the director field around that point, in a local cartesian coordinate system in which \hat{n} is along the Z axis at the origin:

$$\hat{n} = \hat{k} + [(\partial n_x / \partial X)X + (\partial n_x / \partial Y)Y + (\partial n_x / \partial Z)Z] \hat{i} + [(\partial n_y / \partial X)X + (\partial n_y / \partial Y)Y + (\partial n_y / \partial Z)Z] \hat{j}.$$

where \hat{i} , \hat{j} and \hat{k} are dimensionless unit vectors along the X, Y and Z axes respectively. Since the distortion of the director field is assumed to be small, only the first derivatives need be retained in the expansion. Keeping the apolar nature of the director in mind the six first derivatives can be grouped into three sets of curvatures (Fig.4):

$$\text{Splay: } \hat{k} [(\partial n_x / \partial X) + (\partial n_y / \partial Y)] = \hat{n} \text{ div } \hat{n},$$

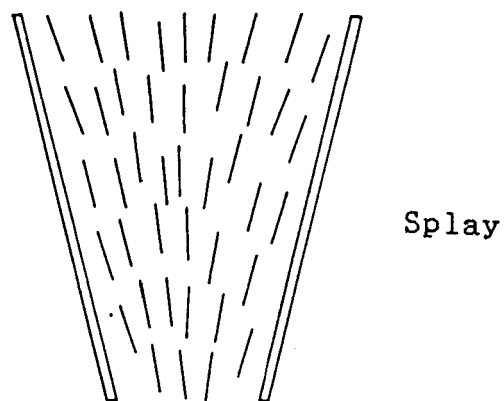
$$\begin{aligned} \text{Twist: } & (\partial n_y / \partial X) - (\partial n_x / \partial Y) = \hat{n} \cdot \text{curl } \hat{n} , \\ \text{Bend: } & - [(\partial n_x / \partial Z)\hat{i} + (\partial n_y / \partial Z)\hat{j}] = \hat{n} \times \text{curl } \hat{n} . \end{aligned}$$

The first and third expressions are polar vectors while the second is a pseudo scalar. Therefore only their squares appear in the expression for the elastic free energy density f^d , which is a scalar quantity. f^d can be expanded in powers of \hat{n} and its gradients. Retaining only those terms that are invariant under the point group symmetry of the nematic and neglecting the surface terms, f^d can be written as:

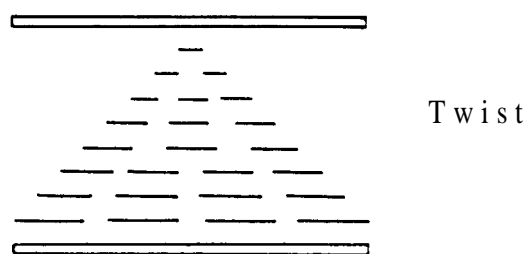
$$\begin{aligned} f^d = & \frac{1}{2} K_1 (\text{div} \hat{n})^2 + \frac{1}{2} K_2 (\hat{n} \cdot \text{curl} \hat{n})^2 + \\ & + \frac{1}{2} K_3 (\hat{n} \times \text{curl} \hat{n})^2 \end{aligned} \quad (1)$$

The K_i are called the Frank elastic constants or the curvature elastic constants. They correspond to the above mentioned three types of deformations possible in a nematic. Since any one of these deformations can be produced independently, the K_i are positive. Usually they are of the order of 10^{-6} dynes.

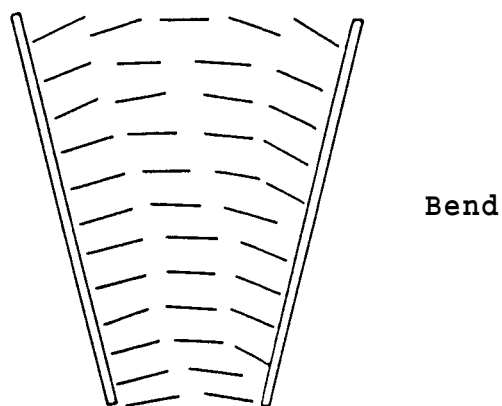
The condition for equilibrium in the absence of external fields can be obtained by minimizing the total distortion energy with respect to all variations of the director $\hat{n}(\vec{r})$ which keep $n^2=1$. Taking into account



Splay



Twist



Bend

Fig.4. The three principal types of deformations in a nematic. (from Ref.1).

this constraint by using the method of Lagrangian undetermined multipliers, the condition for equilibrium is obtained as [2]:

$$h_i^d = - (\partial f^d / \partial n_i) + \partial_j (\partial f^d / \partial g_{ji}) = -c(\vec{r}) n_i \quad (2)$$

where $\partial_j = \partial / \partial x_j$, $g_{ji} = \partial_j n_i$, and $c(\vec{r})$ is an arbitrary function of \vec{r} . \vec{h} is called the molecular field. The above equation states that, in equilibrium, the director, must be parallel to the molecular field at each point. $\vec{h}^d = \vec{h}^s + \vec{h}^t + \vec{h}^b$, where the three parts refer to splay, twist and bend respectively and are given by

$$\begin{aligned} \vec{h}^s &= K_1 \text{grad}(\text{div} \hat{n}) \\ \vec{h}^t &= -K_2 [A \text{curl} \hat{n} + \text{curl}(A \hat{n})] \\ \vec{h}^b &= K_3 [\vec{B} \times \text{curl} \hat{n} + \text{curl}(\hat{n} \times \vec{B})] , \end{aligned} \quad (3)$$

where $A = \hat{n} \cdot \text{curl} \hat{n}$ and $\vec{B} = \hat{n} \times \text{curl}$.

Out of equilibrium, when \hat{n} and \vec{h}^d are not parallel, the director field experiences a torque which tries to make them collinear. The torque density is given by $\vec{\Gamma}^d = \hat{n} \times \vec{h}^d$.

The influence of external fields acting on a nematic can also be described in terms of appropriate molecular

fields. Thus in addition to \vec{h}^d , there will be contributions to the molecular field from each of these external fields. Under such circumstances the equilibrium condition is that \hat{n} must be parallel to the total molecular field \vec{h} .

The elastic constants of a nematic vary considerably with temperature, the variation being approximately proportional to the square of the order parameter S [16]. Twist and bend distortions of the director field are forbidden in the smectic A phase as these alter the layer thickness. As a result K_2 and K_3 are found to diverge strongly close to nematic-smectic A transition points [17,18].

Magnetic Susceptibility

Nematic liquid crystals are usually diamagnetic, being made up of organic molecules. The magnetization \vec{M} induced in the medium by an external field \vec{H} is given by [2],

$$\vec{M} = \chi_{\perp} \vec{H} + (\chi_{\parallel} - \chi_{\perp}) (\hat{n} \cdot \vec{H}) \hat{n} \quad (4)$$

where χ_{\parallel} and χ_{\perp} are the principal diamagnetic susceptibilities parallel and normal to the director, respectively. They are of the order of 10^{-7} cgs electromagnetic

units. The major contribution to the diamagnetic susceptibility comes from the rigid core of the molecules which normally contains aromatic rings. It is energetically favourable for these aromatic rings to orient parallel to an applied magnetic field \vec{H} . As a result nematics made up of rod-like molecules with aromatic cores have a positive diamagnetic anisotropy $\chi_a = \chi_{||} - \chi_{\perp}$ and align with the optic axis parallel to \vec{H} . Due to the same reason, discotic nematics consisting of molecules with aromatic cores have a negative χ_a and orient with their optic axis normal to \vec{H} .

The free energy density of a nematic in a magnetic field is

$$f^m = - \int_0^H \vec{M} \cdot d\vec{H} \quad .$$

Substituting for \vec{M} , we get

$$f^m = -\frac{1}{2} \chi_{\perp} H^2 - \frac{1}{2} \chi_a (\hat{n} \cdot \vec{H})^2$$

The first term is independent of the molecular orientation and hence can be omitted in most problems. The molecular field corresponding to the orientational part of the magnetic free energy can be obtained by minimizing the

free energy as before. It is given by [2]

$$\vec{h}^m = \chi_a (\hat{n} \cdot \vec{H}) \hat{H} \quad (5)$$

If \hat{n} is not parallel to \vec{h}^m , the magnetic torque is given by

$$\vec{\Gamma}^m = \hat{n} \times \vec{h}^m$$

Dielectric Anisotropy

The electric displacement \vec{D} induced in a nematic by an electric field \vec{E} is given by [2]

$$\vec{D} = \epsilon_{\perp} \vec{E} + (\epsilon_{\parallel} - \epsilon_{\perp}) (\hat{n} \cdot \vec{E}) \hat{n} \quad (6)$$

where ϵ_{\parallel} and ϵ_{\perp} are the principal dielectric constants parallel and normal to the director respectively. At low frequencies of the applied field the dielectric anisotropy $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$ may be positive or negative depending on the detailed chemical structure of the constituent molecules. If the molecule is nonpolar or carries a permanent dipole moment parallel to its long axis, ϵ_{\parallel} will be larger than ϵ_{\perp} . In practice large positive values of ϵ_a can be obtained by attaching a strongly polar group at one end of the molecule. If there is a permanent dipole moment which is more or less normal to its long axis we can have $\epsilon_{\parallel} < \epsilon_{\perp}$.

The anisotropy ϵ_a is strongly dependent on temperature.

The free energy density of a nematic in an electric field is given by

$$f^e = -\epsilon_{\perp} E^2 / (8\pi) - \epsilon_a (\hat{n} \cdot \vec{E})^2 / (8\pi)$$

The electric contribution to the molecular field can be obtained by minimizing f^e . It is given by [2]

$$\vec{h}^e = \epsilon_a (\hat{n} \cdot \vec{E}) \vec{E} / (4\pi) \quad (7)$$

The dielectric constants are of course frequency dependent. Both ϵ_{\parallel} and ϵ_{\perp} usually show a Debye type of relaxation at microwave frequencies [19]. Similar relaxation frequencies are also found in the isotropic phase. If the molecule has a nonzero component of electric dipole moment along the long axis, there is an additional relaxation of ϵ_{\parallel} at much lower frequencies, usually in the radio frequency range [20]. This arises because of the strong hindering of the rotation of the molecule about a short axis. For long molecules it is clear that such a rotation is difficult in the nematic phase, and the resulting relaxation frequency is correspondingly small [21].

Electrical Conductivity

The current carriers in a nematic are the impurity ions that are present in the medium. The conductivity can be varied in the range 10^{-11} to 10^{-8} $\text{ohm}^{-1}\text{cm}^{-1}$ by doping the nematic material with suitable ions. The conductivity is anisotropic, with the principal component $\sigma_{||}$ parallel to \hat{n} being larger than the component σ_{\perp} normal to \hat{n} . The positive sign of a , ($= a - a$) can be understood as arising from the greater degree of freedom for the motion of the ions along \hat{n} than for that normal to \hat{n} . For an electric field applied in an arbitrary direction, the current density \mathbf{j} is given by

$$\mathbf{j} = \sigma_{\perp} \vec{E} + \sigma_a (\hat{n} \cdot \vec{E}) \hat{n} \quad (8)$$

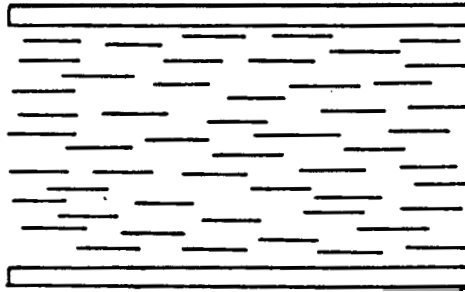
In smectic A and C phases the mobility of the ions in the plane of the layer is larger than that across it. Therefore a is negative in these phases. In some nematics short range smectic-like ordering develops close to smectic (A or C)- nematic transition and a , becomes negative [22].

1.4 ALIGNMENT OF NEMATICS

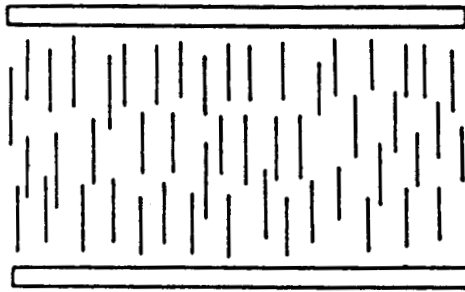
As mentioned earlier, the director \hat{n} can vary gradually from point to point in a nematic. For

experimental purposes it is often necessary to obtain samples with uniform alignment of \hat{n} . Two types of alignments are frequently used: one with the optic axis parallel to the bounding surfaces called planar or homogeneous alignment, and the other with the optic axis normal to them called homeotropic alignment (Fig. 5). Homogeneous alignment can be produced by coating the surfaces with certain polymers followed by unidirectional rubbing [23]. This creates grooves on the surface along the direction of rubbing and, as a result, the nematic is oriented with the director along this direction. Such an alignment can also be produced by vacuum deposition of silicon monoxide at an angle of about 30' to the surface . In this case the shadowing effect produces grooves on the surface [23]. Homeotropic alignment is usually produced by coating the surface with certain polymers which have chemical groups that can attach themselves to the surface and long chains that stick out of the surface. In many cases very clean glass plates give a homeotropic alignment even without any further treatment.

Due to the diamagnetic and dielectric anisotropy of nematics, an external magnetic or electric field can also be used to align the director in the medium. Depending



(a)



(b)

Fig.5. Schematic representation of (a) homogeneous and (b) homeotropic alignments.

on the sign of the corresponding anisotropy, $\hat{\mathbf{n}}$ is aligned either parallel or normal to the field direction. In many experiments external magnetic and electric fields are used to induce distortions in a surface-aligned monodomain sample (see Chapter 7). These bulk distortions can influence the orientation of $\hat{\mathbf{n}}$ at the surface if the surface anchoring is weak. Under such conditions, the surface terms in the free energy should be taken into account to determine the equilibrium state. On the other hand, if the surface anchoring is strong, these bulk distortions cannot influence the orientation of $\hat{\mathbf{n}}$ at the surface and the surface terms in the free energy can be neglected.

1.5 HYDRODYNAMICS OF NEMATICS

Due to the absence of any positional ordering of the molecules in the medium, a nematic can flow with ease. The flow properties are, however, very complex due to the orientational ordering of the molecules. The translational motion of the molecules is coupled to their orientational motion. As a result, a flow in the medium can disturb the alignment of the molecules under many circumstances. A continuum theory of the flow properties of nematics was developed by Ericksen [24] and Leslie [25], a brief account of which is given below. We find it convenient to use the notation of de Gennes [2] as it is the one widely

used in the literature on electrohydrodynamic problems.

The dynamical state of a flowing isotropic liquid is described by the velocity field $\vec{v}(\vec{r})$. Assuming the fluid to be incompressible, its equation of motion is given by the Navier - Stokes equation [26],

$$\rho \, dv_j / dt = \partial_i \sigma_{ij} \quad ,$$

where ρ is the density, $\partial_i \equiv \partial / \partial x_i$, $\sigma_{ij} = -p \delta_{ij} + \sigma'_{ij}$, is the stress tensor, p the pressure and σ'_{ij} the viscous stress tensor. σ'_{ij} describes the irreversible transfer of momentum in the fluid and an expression for it can be found by considering the entropy source due to all the frictional processes in the medium [27]. Since in an isotropic fluid only a relative motion of its different parts can give rise to internal friction, σ'_{ij} depends only on the velocity gradients and is given by

$$\sigma'_{ij} = \eta \, A_{ij}$$

where $A_{ij} = \frac{1}{2} (\partial_j v_i + \partial_i v_j)$ and η is the viscosity coefficient.

As mentioned earlier, the translational and rotational degrees of freedom in a nematic are coupled in most but

not all situations. This necessitates the choice of an additional variable describing the local alignment of the molecules to describe its dynamical state. This is done in terms of the director $\hat{\mathbf{n}}(\vec{\mathbf{r}})$. The equations of motion of these two variables, viz, $\vec{\mathbf{v}}(\vec{\mathbf{r}})$ and $\hat{\mathbf{n}}(\vec{\mathbf{r}})$ can be obtained by determining the entropy source due to all frictional processes in the medium. Assuming these processes to be isothermal, it is given by

$$T \dot{s} = -(d/dt) \int [\frac{1}{2} \rho v^2 + f^o + f^d + f^F] dV \quad (9)$$

where T is the temperature, s the entropy, f^o an internal free energy density, f^d the distortion free energy density and f^F the free energy density due to an external electric or magnetic field. The acceleration equation for the fluid can be written as

$$\rho dv_j/dt = \partial_i \sigma_{ij} \quad (10)$$

where σ_{ij} is the stress tensor. Substituting Eq.(10) in Eq.(9) and integrating by parts, we get the contribution of the kinetic energy to the entropy source to be

$$-(d/dt) \int (\frac{1}{2} \rho v^2) dV = \int \sigma_{ij} \partial_i v_j dV \quad (11)$$

A change in f^F can arise from small changes in $\hat{\mathbf{n}}$ at

each point in the medium. This is given by

$$\delta f^F = \left[\left(\frac{\partial f^F}{\partial n_i} \right) - \partial_j \left(\frac{\partial f^F}{\partial g_{ji}} \right) \right] \delta n_i = - h_i^F \delta n_i$$

where \vec{h}^F is the corresponding molecular field. Therefore the contribution of f^F to the entropy source is given by

$$-(d/dt) \int f^F dV = \int \vec{h}^F \cdot \vec{\hat{n}} dV \quad (12)$$

A change in f^d can be split up into two parts. The first part results from small changes in \hat{n} at each point in the medium and as in the case of the external field it is

$$\delta f_i^d = - h_i^d \delta n_i$$

where \vec{h}^d is the molecular field due to the distortion free energy.

The second part arises from small displacements of \hat{n} which conserve its orientation. Considering a displacement of the form $\vec{r}' = \vec{r} + \vec{u}(\vec{r})$, the change in the free energy is given by

$$\delta f_2^d = \left(\frac{\partial f^d}{\partial g_{3i}} \right) \left[\left(\frac{\partial n'_i}{\partial x'_j} \right) - \left(\frac{\partial n_i}{\partial x_j} \right) \right]$$

It can be easily shown that

$$\left(\frac{\partial n_i'}{\partial x_j'} \right) - \left(\frac{\partial n_i}{\partial x_j} \right) = - \left(\frac{\partial_k n_i}{\partial x_j} \right) \left(\frac{\partial_j u_k}{\partial x_j} \right)$$

Therefore the change in the free energy can be written as

$$\delta f^d = \sigma_{jk}^d \partial_j u_k$$

where $\sigma_{jk}^d = - \left(\frac{\partial f^d}{\partial g_{ji}} \right) \frac{\partial_k n_i}{\partial x_j}$ and $g_{ji} = \frac{\partial_j n_i}{\partial x_j}$.

σ_{ij}^d is called the distortion or elastic stress tensor. It is not symmetric in general. This asymmetry arises from the fact that a displacement corresponding to a pure rotation of the centres of mass of the molecules which conserves the orientation of \hat{n} usually alters the distortion energy. However if the elastic constants are equal, f^d becomes invariant with respect to rotations in \vec{u} space and in \hat{n} space separately and σ_{ij}^d becomes symmetric.

If the medium is incompressible, then a displacement of \hat{n} results in a change in f^o given by

$$\delta f^o = -p \delta_{ij} \partial_i u_j$$

Hence the variation in the total free energy due to small displacements of \hat{n} which conserve its orientation is

$$\delta(f_2^d + f^o) = \sigma_{ij}^e \partial_i u_j$$

where $\sigma_{ij}^e = \sigma_{ij}^d - p \delta_{ij}$ is the Ericksen or equilibrium stress tensor. Therefore the contribution of f^o , f^d and f^F to the entropy source is given by

$$-(d/dt) \int [f^o + f^d + f^F] dV = \int [-\sigma_{ij}^e \partial_i v_j + \vec{h} \cdot \vec{\dot{n}}] dV \quad (13)$$

where $\vec{h} = \vec{h}^d + \vec{h}^F$ is the total molecular field.

The total entropy source can now be written as

$$\tau \dot{s} = \int [(\sigma_{ij} - \sigma_{ij}^e) \partial_i v_j + \vec{h} \cdot \vec{\dot{n}}] dV \quad (14)$$

where $\vec{\dot{n}}$ is the material derivative of \hat{n} . The difference $\bar{a} - \bar{a}^e \equiv \bar{a}'$ is the viscous stress tensor. \bar{a}' is not symmetric in general. The antisymmetric part can be expressed in terms of a vector $\vec{\Gamma}$ defined as

$$\Gamma_z = \sigma'_{yx} - \sigma'_{xy}, \text{ etc.} \quad (15)$$

From angular momentum conservation equation it can be shown that

$$\vec{\Gamma} = \hat{n} \times \vec{h} \quad (16)$$

$\vec{\Gamma}$ is therefore the torque density 'exerted' by the internal degree of freedom \hat{n} on the flow. Since \hat{n} and \vec{h} are collinear in equilibrium, this torque is nonzero only when the system is not in equilibrium. Thus the asymmetry of \vec{a} reflects the presence of these bulk torques.

Using Eq.(14) the expression for the entropy source can be further simplified as

$$\tau \dot{s} = \int [\bar{\bar{A}} : \bar{\bar{\sigma}}^s + \vec{h} \cdot \vec{N}] dV \quad (17)$$

where $\bar{\bar{\sigma}}^s$ is the symmetric part of $\bar{\bar{\sigma}}$,
 $\vec{N} = \dot{\vec{h}} - \vec{w} \times \hat{n}$ and $\vec{w} = \frac{1}{2} \text{curl } \vec{v}$.

\vec{N} represents the rate of change of the director orientation with respect to the background fluid. Eq.(17) clearly shows that there are two contributions to the entropy source. The first term represents the losses due to conventional viscosity effects and the second term gives the losses associated with a rotation of \hat{n} with respect to the background fluid. Each contribution to the entropy source can be written as the product of a 'flux' and the conjugate 'force' [27]. Assuming that the fluxes are weak, the forces can be expressed as linear functions

of the fluxes. Taking A_{ij} and N_i as the fluxes, we have

$$\sigma_{ij}^s = L_{ijkl} A_{kl} + M_{ijk} N_k,$$

$$h_l = M'_{ijl} A_{ij} + P_{lk} N_k.$$

By the Onsager reciprocity relations M and M' must be identical. The structure of the matrices L, M and P must be compatible with the local symmetry of the nematic. Assuming the nematic to be incompressible, the most general forms of the stress tensor and the molecular field are

$$\begin{aligned} \sigma'_{ij} = & a_1 n_i n_j n_k n_l A_{kl} + a_4 A_{ij} + a_5 n_i n_l A_{lj} \\ & + a_6 n_l n_j A_{il} + a_2 n_i N_j + a_3 n_j N_i \end{aligned} \quad (18)$$

and

$$h_i = \gamma_1 N_i + \gamma_2 n_j A_{ij} \quad (19)$$

where $\gamma_1 = a_3 - a_2$ and $\gamma_2 = a_3 + a_4$.

The coefficients a_i are called the Leslie viscosity coefficients and are usually of the order of 10^{-1} to 1 poise. Using the Onsager reciprocity relations Parodi [28] showed that the a_i are related by the relation

$$a_2 + a_3 = a_6 - a_5.$$

Hence the dynamics of an incompressible nematic involves five independent viscosity coefficients. The meaning of the different terms in $\bar{\sigma}$ ' is illustrated in Fig.6 [29]. The α_1 term corresponds to the usual viscosity of an isotropic fluid. The α_2 term is symmetric and expresses the stretching effect of an irrotational flow. The α_3 and α_4 terms are asymmetric and express the torques exerted on \hat{n} by an irrotational flow. The α_5 and α_6 terms are also asymmetric and express the torques due to a rotational flow.

Thus in the Ericksen-Leslie theory the equations describing a flowing nematic are:

1) the acceleration equation-

$$\rho \, dv_j / dt = \partial_i \sigma_{ij} ,$$

where $\bar{\sigma} = \bar{\sigma}^e + \bar{\sigma}'$. When the dependence on the director and its gradients is neglected, this equation reduces to the Navier-Stokes equation for an isotropic fluid.

2) the equation $h_i = \gamma_1 N_i + \gamma_2 n_j A_{ij}$, which gives the rate of change of the director in terms of the velocity

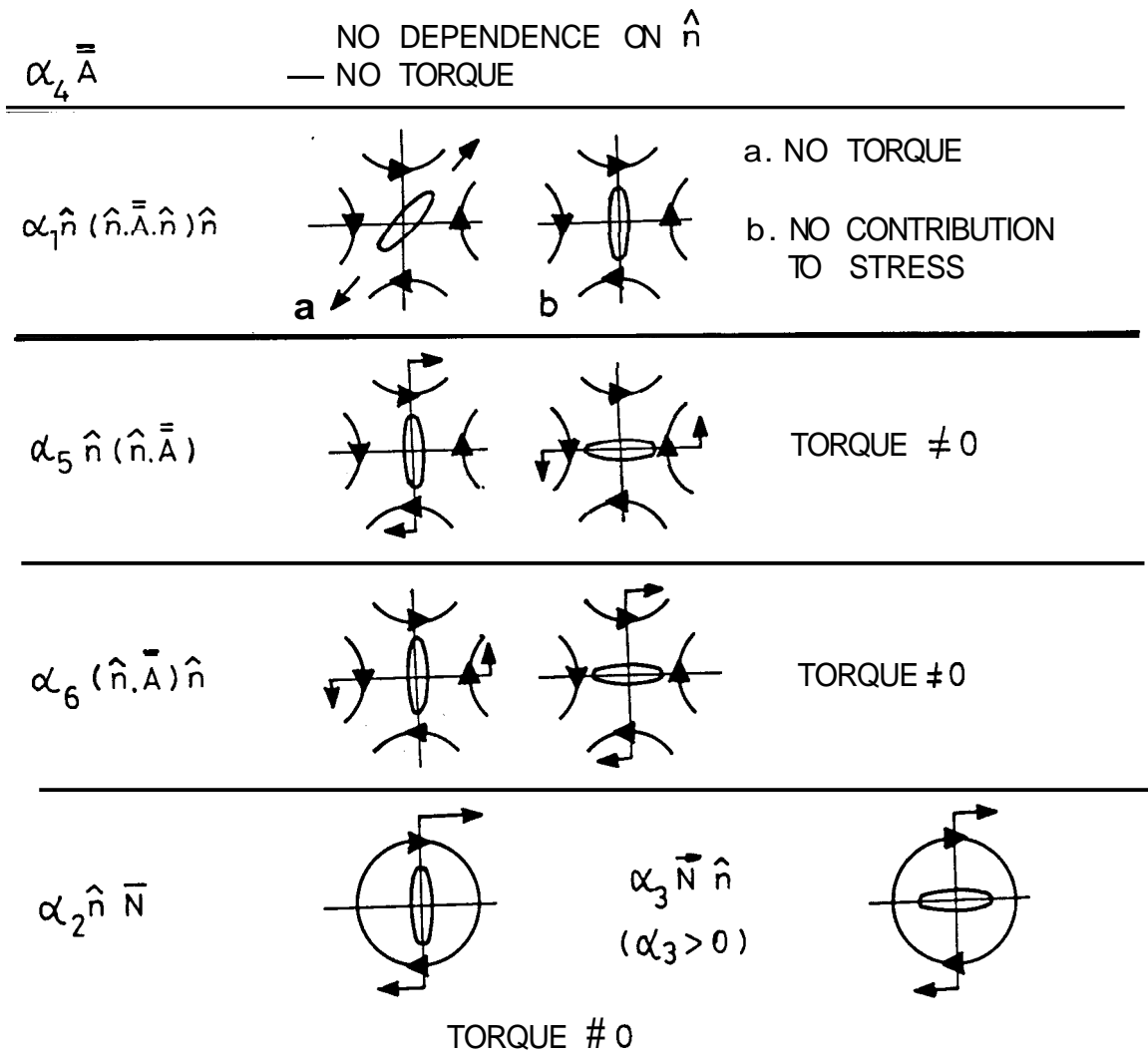


Fig.6. The meaning of the different terms in the viscous tensor. (From Ref.29).

gradients and the molecular field.

The coupling between orientation and flow in nematics leads to many interesting flow properties. One such is the so-called nematic Hall-effect [30]. Consider the Poiseuille flow of a nematic between two horizontal planes as shown in Fig.7 . If the director is oriented at an angle to the flow direction using an external magnetic field applied at an angle ϕ to the flow direction, then a transverse pressure gradient is found to develop in the medium. This can be understood in terms of the Ericksen-Leslie theory. Using the Leslie stress tensor $\bar{\mathbf{a}}'$, the viscous forces acting in the fluid are found to be

$$F_y = (\eta_a \cos^2 \phi + \eta_b \sin^2 \phi) (ds/dz) = \eta(\phi) ds/dz$$

and

$$F_x = (\eta_b - \eta_a) \sin \phi \cos \phi ds/dz.$$

where $\eta_a = \frac{1}{2} \alpha_4$, $\eta_b = \frac{1}{2} (\alpha_3 + \alpha_5 + \alpha_6)$ and $s = (dv_y / dz)$ is the local shear rate. The presence of F_x leads to a transverse flow in the medium. In the steady state F_x and F_y must be balanced by pressure gradients. Therefore,

$$(\partial p / \partial x) / (\partial p / \partial y) = [(\eta_b - \eta_a) / \eta(\phi)] \sin \phi \cos \phi.$$

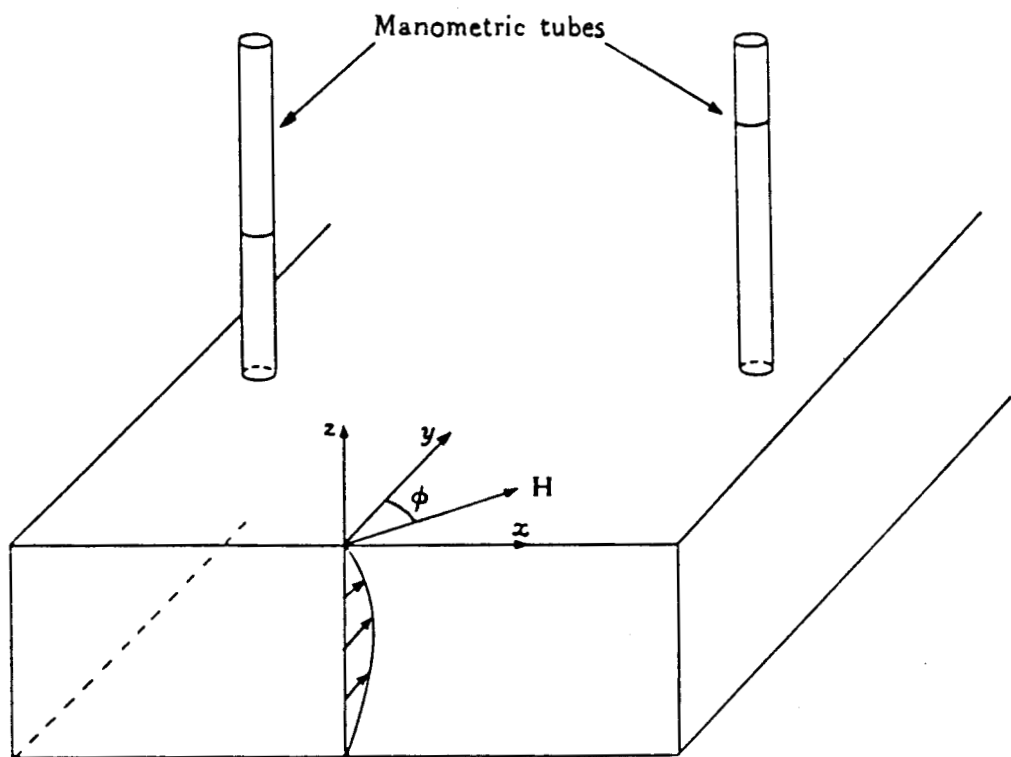


Fig.7. Poiseuille flow under a strong oblique magnetic field. In the absence of H the flow is along y . (from Ref. 2).

It is clear from the above equation that when the initial flow is either along the director or normal to **it**, there is no transverse flow. Otherwise the director tends to deflect the initial flow giving rise to a transverse flow in the medium.

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