

# THEORY OF LIQUID CRYSTALS-MODELLING ELECTRIC FIELDS

Nigel Mottram  
Department of Mathematics



*The Static and Dynamic Continuum Theory of Liquid Crystals*  
by I. W. Stewart, Taylor & Francis (Oct 2002).

## Lecture 1: Theory of Liquid Crystals

- Theoretical description of liquid crystals: macroscopic quantities
- Energy: elasticity, surface interactions, magnetic fields
- Static theory of liquid crystals: nematic examples
- (Dynamic theory of liquid crystals: director rotation and flow)

## Theoretical description of liquid crystals

In order to construct a mathematical description of the many liquid crystalline phases (nematic, smectic A, smectic C etc.) we need to find quantities which specify the amount and type of order within the phase.

In a nematic we need to describe the amount and direction of orientational order.

In a smectic phase we need to describe the density changes due to the layer structure **and** the amount and direction of orientational order in each layer.

We could consider the liquid crystal on an **atomistic** level.

In this case the position of each atom in each molecule is considered.

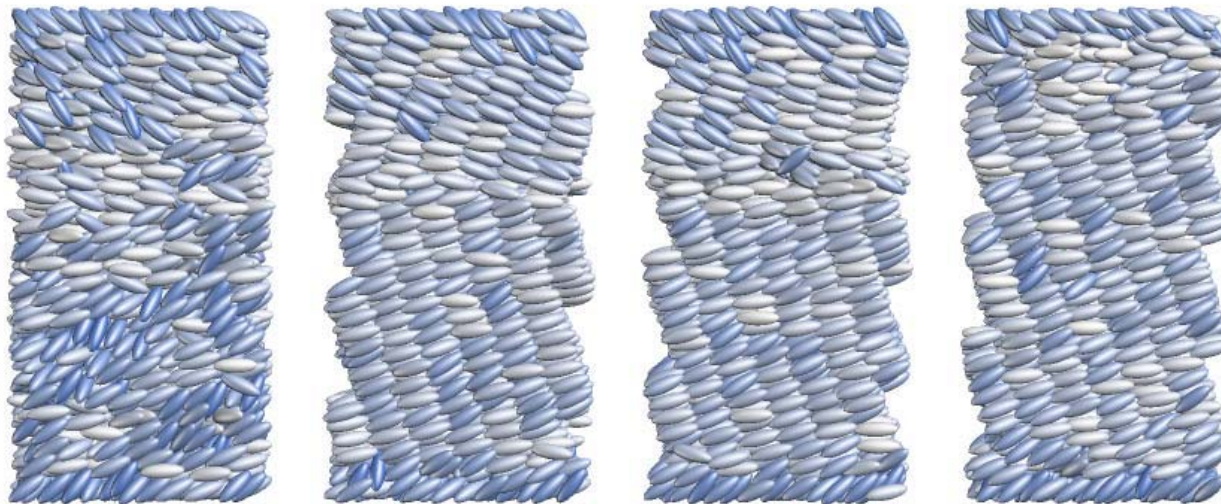
For a typical liquid crystal device the number of variables is then very large.

A region of material of size,  $1\text{mm} \times 1\text{mm} \times 1\mu\text{m}$  could contain  $10^{17}$  molecules, each of which could contain  $10^2$  atoms so there would be  $10^{19}$  variables!

Atomistic modelling has been carried out with around 100 molecules in order to investigate the molecule-molecule or molecule-substrate interaction.

Even if we consider a simplified form of the molecule (a rod or ellipsoid) we still need  $10^{17}$  molecules for a reasonably sized pixel.

Using up to  $10^6$  molecules, these **molecular** simulations have shown certain bulk properties of the LC and in recent years have been used to model extremely thin cells.

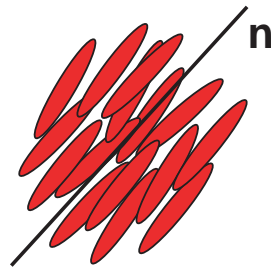


In order to realistically model an LC device (or at least each pixel) it is common to use a **macroscopic theory**.

In a macroscopic theory the liquid crystal is described in terms of *macroscopic* variables.

These variables are properties of a group of molecules. The molecular properties are averaged over an tiny region of liquid crystal.

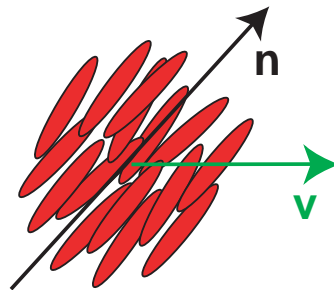
For instance, the **director** is not a molecular direction but the average molecular direction in an area around a point in the LC.



Other macroscopic variables such as the density of an LC are only really defined in terms of a collection of molecules in an LC region.

Each phase of a material will be described by a set of macroscopic variables.

For instance, a nematic may be described by the director  $\mathbf{n}$ , a vector quantity, and if the LC is flowing, the fluid velocity  $\mathbf{v}$ , another vector quantity.



The director and velocity may be different in different parts of the cell so they depend on the position vector  $\mathbf{r} = (x, y, z)$  and possibly time  $t$ .

We write  $\mathbf{n} = \mathbf{n}(x, y, z, t)$ ,  $\mathbf{v} = \mathbf{v}(x, y, z, t)$

## Continuum theory

Given certain influences on the liquid crystal we would like to find out how the macroscopic variables vary through the material.

Does the director change direction from one place in the LC to another?

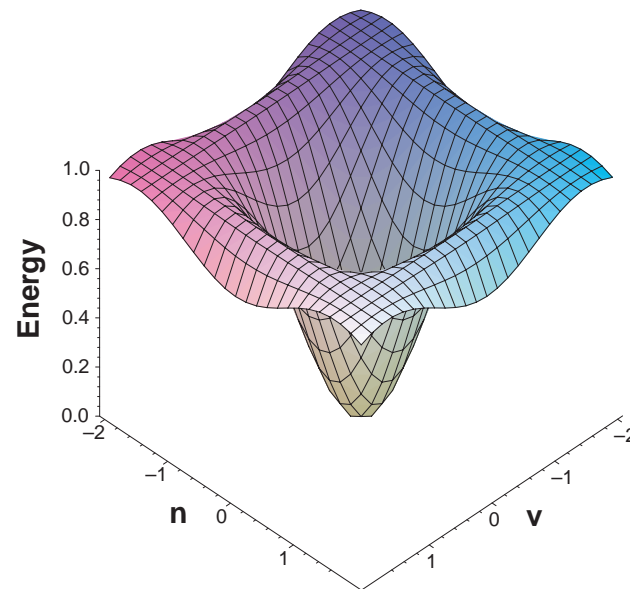
Is the fluid velocity larger or smaller at the centre of a cell?

A continuum theory treats the material as a continuum: rather than consider discrete molecules the macroscopic averages are assumed to be continuous within the material.

With this assumed continuity we can define gradients of variables,  $\frac{\partial \mathbf{n}}{\partial x}$ , i.e. the change of the director in the  $x$  direction.

The internal energy of the material can be separated into various terms: elastic energy, electrostatic energy, surface interaction energy etc.

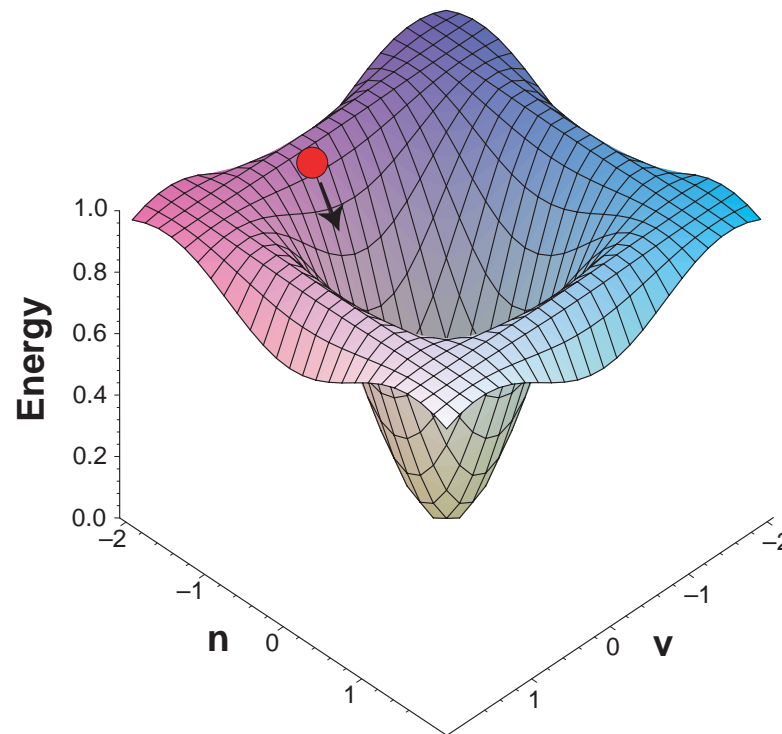
In a **static** continuum theory we write these energies in terms of the macroscopic variables and assume that, in equilibrium, the system will be in an energy minimum.



We must therefore find the values of the macroscopic variables (or the functional forms if they vary in space) which minimise the energy.

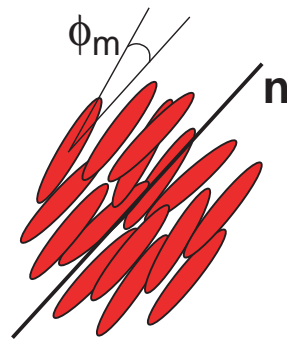
In a **dynamic** continuum theory energy may dissipate as the fluid flows, the director rotates and through viscous forces.

The macroscopic variables may change in time as the system tries to reach the static equilibrium state or external influences (e.g. an electric field) force the system out of the energy minimum.



In a nematic liquid crystal there is orientational ordering but no positional ordering (the molecular centres of mass are randomly positioned)

It is possible to define a macroscopic quantity which describes this orientational ordering, the **director**  $\mathbf{n}$ , of unit length.

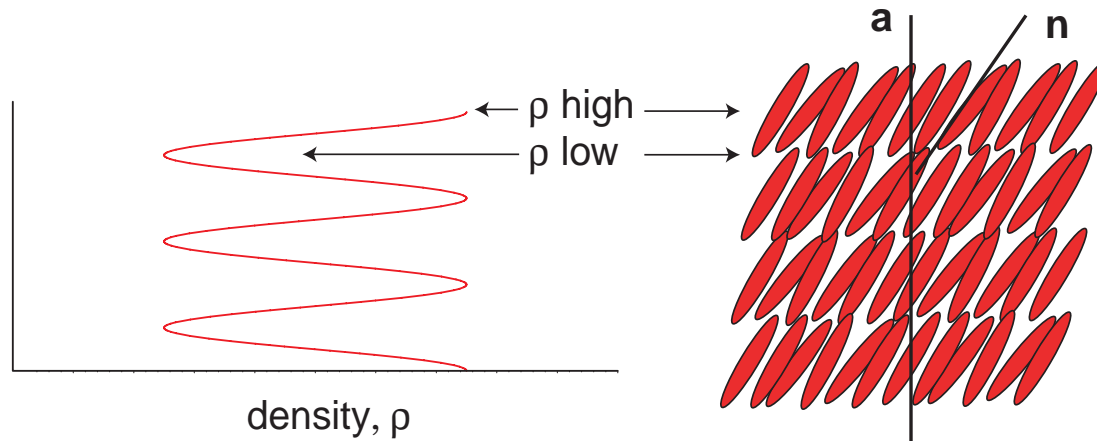


However, we can define both the direction of the orientational order and the **magnitude** of orientation.

How aligned with  $\mathbf{n}$  are the molecules in the area?

If  $\phi_m$  is the angle between a molecule and the director then the **scalar order parameter** is  $S = \frac{1}{2} \langle 3 \cos^2(\phi_m) - 1 \rangle$  where  $\langle \rangle$  denotes the local average.

In the smectic phase the director  $\mathbf{n}$  and layer normal direction  $\mathbf{a}$  must be specified in order to determine the configuration.



We can also define the nematic-like scalar order parameter  $S$  and the density function  $\rho$  which is related to the probability of finding a molecular centre of mass at a certain point.

$\rho$  is high at a point in the middle of a layer and low in the inter-layer region.

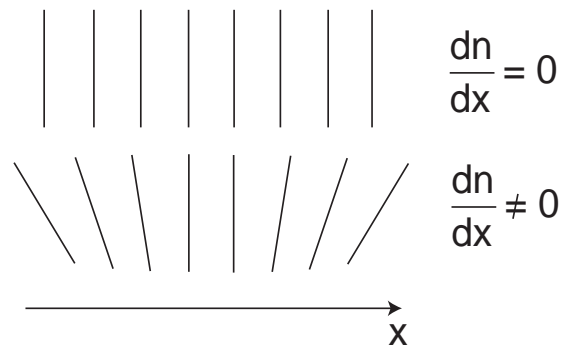
In the theories used to describe display devices it is often assumed that the nematic scalar order parameter is constant (it only changes significantly, close to defects) and in smectics that the layer structure is fixed so that  $\mathbf{a}$  and  $\rho$  are constant functions.

The relevant variable for a display device is then simply the director  $\mathbf{n}$  although in some cases the fluid velocity  $\mathbf{v}$  should be included for more accurate results.

To construct a continuum theory of liquid crystals we must now consider the various energy contributions and how they depend on the director.

## Elasticity

Since the LC molecules prefer to align, on average, in a fixed direction, any distortion of the director structure will be disfavoured and will increase the internal energy of the system.



The **elastic** energy of the LC will therefore be a function of the director  $\mathbf{n}$  and gradients of  $\mathbf{n}$ .

To find this function we use certain assumptions about the LC system which depend on the symmetry of the phase.

For a nematic LC we assume,

- the director is “headless” so that  $\mathbf{n} \rightarrow -\mathbf{n}$  leaves the energy unchanged.
- any translation of the system ( $x \rightarrow x + 1$ ) leaves the energy unchanged.
- any rigid rotation of the system leaves the energy unchanged (looking at the LC from a different angle shouldn't change the energy).
- there are only small distortions present so that we may neglect terms smaller than  $\left(\frac{\partial \mathbf{n}}{\partial x}\right)^2$ .

which leads to the energy density

$$w_d = \frac{1}{2}K_1 (\nabla \cdot \mathbf{n})^2 + \frac{1}{2}K_2 (\mathbf{n} \cdot \nabla \times \mathbf{n} - q)^2 + \frac{1}{2}K_3 (\mathbf{n} \times \nabla \times \mathbf{n})^2 + \frac{1}{2}(K_2 + K_4) \nabla \cdot ((\mathbf{n} \cdot \nabla) \mathbf{n} - (\nabla \cdot \mathbf{n}) \mathbf{n})$$

where  $\nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z}\right)$  is shorthand for the derivatives in each direction and the  $\cdot$  and  $\times$  are types of products of vectors.

It is relatively straightforward to see that:

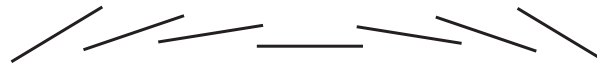
- the first distortion term  $\nabla \cdot \mathbf{n}$  is associated with splaying of the director,
- the second term  $\mathbf{n} \cdot \nabla \times \mathbf{n}$  is associated with twisting of the director,
- the third term  $\mathbf{n} \times \nabla \times \mathbf{n}$  is associated with bending of the director,
- and the fourth term is associated with a distortion in two directions (a saddle-splay term).



splay



twist



bend

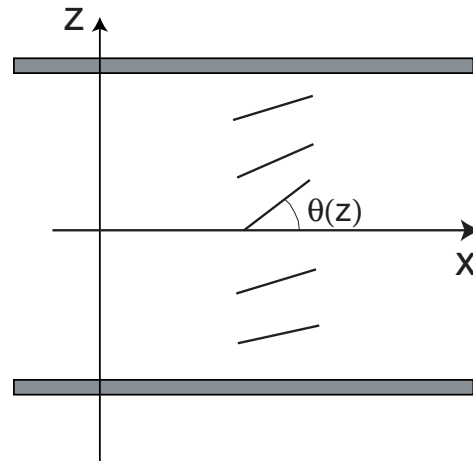
$$w_d = \frac{1}{2}K_1 (\nabla \cdot \mathbf{n})^2 + \frac{1}{2}K_2 (\mathbf{n} \cdot \nabla \times \mathbf{n} - q)^2 + \frac{1}{2}K_3 (\mathbf{n} \times \nabla \times \mathbf{n})^2 + \frac{1}{2}(K_2 + K_4) \nabla \cdot ((\mathbf{n} \cdot \nabla) \mathbf{n} - (\nabla \cdot \mathbf{n}) \mathbf{n})$$

The  $K$  constants are specific to each LC material, are temperature dependent and are called the Frank elastic constants.

The constant  $q$  is the natural twist of the material which is nonzero only in chiral nematics. It can also be shown that  $(K_2 + K_4) = 0$  in a chiral nematic.

It is common (but not very accurate) to use the “one constant approximation” which asserts that  $K_1 = K_2 = K_3 = K$  and  $K_4 = 0$  (some authors write  $K_2 + K_4 = 0$  instead).

Example: Simple nematic cell



If the director lies in the  $xz$ -plane and the angle between the director and the  $x$ -axis is  $\theta$  which varies only in the  $z$  direction, perpendicular to the substrates then the elastic energy is

$$w_d = \frac{1}{2} K \left( \frac{d\theta}{dz} \right)^2$$

If  $\theta$  is constant there is no elastic energy. If  $\theta$  varies through the cell the elastic energy is nonzero.

## Surfaces

In most LC devices the glass or plastic substrates within which the LC material is sandwiched are treated to provide some form of alignment of the director.

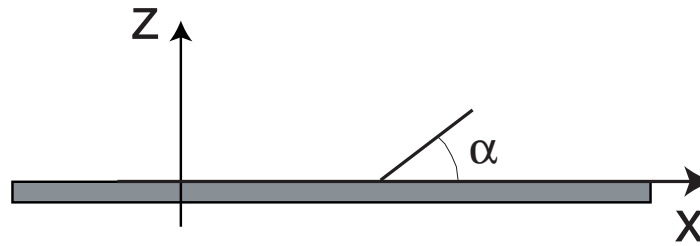
The main forms of alignment are

- strong anchoring: the alignment layer at the substrate is strong enough to fix the LC molecules there. The director is therefore fixed to be a specific value  $\mathbf{n} = \mathbf{n}_s$  at the substrate.
- weak anchoring: the alignment method defines a fixed direction, a preferred direction,  $\mathbf{n}_s$ . The director at the surface may take a different value from  $\mathbf{n}_s$  but this will cost energy.

Mathematically, strong anchoring gives a fixed boundary condition,  $\mathbf{n} = \mathbf{n}_s$ .

For weak anchoring we specify a surface energy  $w_s = W_s(1 - (\mathbf{n} \cdot \mathbf{n}_s)^2)$  (Rapini-Papoular) which is added to the bulk energy. A solution for  $\mathbf{n}$  should then minimise the total energy.

Example: Simple nematic cell



If the alignment layer prefers the director with a pretilt angle  $\alpha$  then  $\mathbf{n}_s = (\cos \alpha, 0, \sin \alpha)$ .

A strong anchoring condition would be  $\theta = \alpha$  at  $z = 0$ .

With weak anchoring the surface energy is  $w_s = W_s(1 - (\mathbf{n} \cdot \mathbf{n}_s)^2) = W_s \sin^2(\theta - \alpha)$ .

When  $\alpha = 0$  the director prefers to lie parallel to the substrate. This type of anchoring is termed homogeneous or planar anchoring.

When  $\alpha = \pi/2$  the director prefers to lie perpendicular to the substrate. This type of anchoring is termed homeotropic anchoring

## Magnetic fields

A liquid crystal molecule placed in a magnetic field  $\mathbf{H}$  will distort that field inducing a magnetisation  $\mathbf{M}$  and increasing the magnetic energy.

It will distort the magnetic field by different amounts depending of the orientation of the molecule.

- If  $\mathbf{n}$  is parallel to  $\mathbf{H}$  the magnetisation is  $\mathbf{M} = \chi_{||} \mathbf{H}$
- If  $\mathbf{n}$  is perpendicular to  $\mathbf{H}$  the magnetisation is  $\mathbf{M} = \chi_{\perp} \mathbf{H}$

For a general director the magnetisation is  $\mathbf{M} = \chi_{\perp} \mathbf{H} + \chi_a (\mathbf{n} \cdot \mathbf{H}) \mathbf{n}$ .

The magnetic susceptibilities  $\chi_{||}$  and  $\chi_{\perp}$  are usually negative whilst the diamagnetic anisotropy  $\chi_a = \chi_{||} - \chi_{\perp}$  is positive.

The magnetic energy density is

$$\begin{aligned}w_m &= -\frac{1}{2}\mathbf{M}\cdot\mathbf{H} \\ &= -\frac{1}{2}\chi_{\perp}\mathbf{H}^2 - \frac{1}{2}\chi_a(\mathbf{n}\cdot\mathbf{H})^2\end{aligned}$$

To be accurate, for a specific director configuration, the magnetic field  $\mathbf{H}$  should be calculated from Maxwell's equations for the field and the magnetic induction  $\mathbf{B}$

$$\nabla \times \mathbf{H} = 0$$

$$\nabla \cdot \mathbf{B} = 0$$

The magnetic induction is

$$\begin{aligned}\mathbf{B} &= \mu_0(\mathbf{H} + \mathbf{M}) \\ &= \mu_0((1 + \chi_{\perp})\mathbf{H} + \chi_a(\mathbf{n} \cdot \mathbf{H})\mathbf{n})\end{aligned}$$

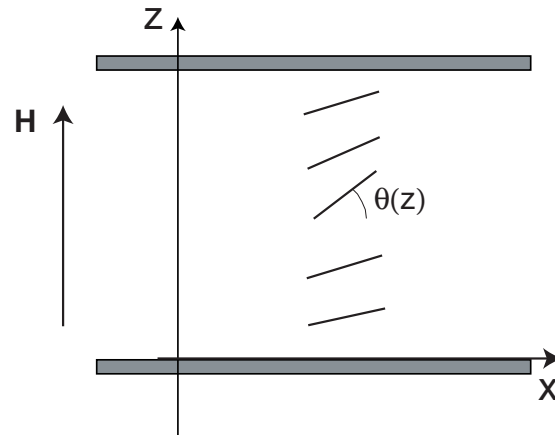
where  $\mu_0$  is the permeability of free space.

Since  $\chi_a$  is usually small (approximately  $10^{-6}$ ) compared with  $1 + \chi_{\perp}$  then we assume  $\mathbf{B}$  is directly proportional to  $\mathbf{H}$  and Maxwell's equations become

$$\begin{aligned}\nabla \times \mathbf{H} &= 0 \\ \nabla \cdot \mathbf{H} &= 0\end{aligned}$$

which is solved by setting  $\mathbf{H}$  to be constant.

Example: Simple nematic cell



With a constant magnetic field  $\mathbf{H} = (0, 0, H)$  in the  $z$  direction and the director  $\mathbf{n} = (\cos \theta, 0, \sin \theta)$  then the magnetic energy density is

$$w_m = -\frac{1}{2}\chi_{\perp}H^2 - \frac{1}{2}\chi_a H^2 \sin^2 \theta$$

If  $\chi_a > 0$  the lowest magnetic energy state exists when  $\theta = \pi/2$  so the director aligns with the field.

If  $\chi_a < 0$  the lowest magnetic energy state exists when  $\theta = 0$  so the director aligns perpendicular to the field.

## Magnetic units

Care must be taken when specifying the units of the magnetic field, induction and magnetisation.

In cgs units the magnetic field  $\mathbf{H}$  and magnetisation  $\mathbf{M}$  are measured in oersted (Oe) and the magnetic induction  $\mathbf{B}$  is measured in gauss (G).

But since  $\mu_0 = 1$  (unitless) in cgs units and  $\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M})$ , a magnetic field is sometimes measured in gauss.

In SI units the magnetic field  $\mathbf{H}$  and magnetisation  $\mathbf{M}$  are measured in amperes per metre (A/m) and the magnetic induction is measured in Tesla (T).

In SI units  $\mu_0 = 4\pi \times 10^{-7}$  H/m so care must be taken when changing from field to induction units.

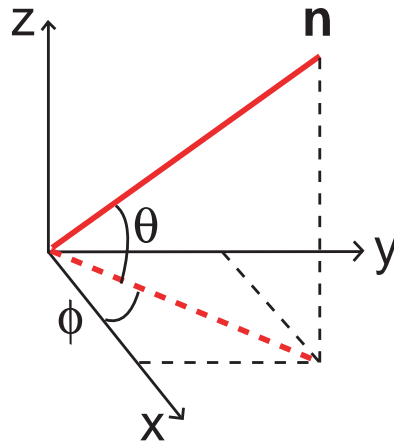
## Energy minimisation

Each energy contribution depends on  $\mathbf{n}_i$  and derivatives  $\frac{\partial \mathbf{n}_i}{\partial x}$ ,  $\frac{\partial \mathbf{n}_i}{\partial y}$ ,  $\frac{\partial \mathbf{n}_i}{\partial z}$ .

Or since  $\mathbf{n}$  can be written in terms of two angles  $\theta$  and  $\phi$  so that

$$\mathbf{n} = (\cos \theta \cos \phi, \sin \phi, \sin \theta \cos \phi)$$

the energy can be written in terms of  $\theta$ ,  $\phi$  and all derivatives such as  $\frac{\partial \theta}{\partial x}$ .



Given the energy density, which is the sum of elastic, surface and magnetic energy densities, how do we find the director structure  $\mathbf{n}(x, y, z)$  (or  $\theta(x, y, z)$ ,  $\phi(x, y, z)$ ) which minimises the energy.

If the total energy is

$$F = \int_V w_d + w_m \, dv + \int_S w_s \, ds$$

where  $V$  is the volume occupied by the LC material and  $S$  is the boundary of  $V$ ,

then a minimum director configuration given by  $\theta(x, y, z)$ ,  $\phi(x, y, z)$  is found by solving the Euler-Lagrange equations,

$$0 = \frac{\partial}{\partial x} \left( \frac{\partial w}{\partial \frac{\partial \theta}{\partial x}} \right) + \frac{\partial}{\partial y} \left( \frac{\partial w}{\partial \frac{\partial \theta}{\partial y}} \right) + \frac{\partial}{\partial z} \left( \frac{\partial w}{\partial \frac{\partial \theta}{\partial z}} \right) - \frac{\partial w}{\partial \theta}$$

$$0 = \frac{\partial}{\partial x} \left( \frac{\partial w}{\partial \frac{\partial \phi}{\partial x}} \right) + \frac{\partial}{\partial y} \left( \frac{\partial w}{\partial \frac{\partial \phi}{\partial y}} \right) + \frac{\partial}{\partial z} \left( \frac{\partial w}{\partial \frac{\partial \phi}{\partial z}} \right) - \frac{\partial w}{\partial \phi}$$

where  $w = w_d + w_m$ .

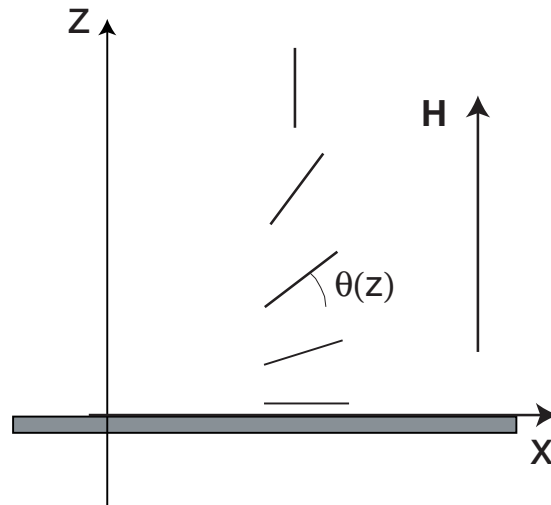
with the weak anchoring boundary condition

$$0 = \nu_1 \frac{\partial w}{\partial \frac{\partial \theta}{\partial x}} + \nu_2 \frac{\partial w}{\partial \frac{\partial \theta}{\partial y}} + \nu_3 \frac{\partial w}{\partial \frac{\partial \theta}{\partial z}} + \frac{\partial w_s}{\partial \theta}$$
$$0 = \nu_1 \frac{\partial w}{\partial \frac{\partial \phi}{\partial x}} + \nu_2 \frac{\partial w}{\partial \frac{\partial \phi}{\partial y}} + \nu_3 \frac{\partial w}{\partial \frac{\partial \phi}{\partial z}} + \frac{\partial w_s}{\partial \phi}$$

where  $\nu = (\nu_1, \nu_2, \nu_3)$  is the substrate normal direction.

If strong anchoring is present the above boundary condition is replaced by  $\theta = \alpha_\theta$  and  $\phi = \alpha_\phi$  on the boundary.

Example: Magnetic coherence length



At the boundary there is strong anchoring so that  $\theta = 0$  at  $z = 0$ .

With the one constant approximation the energy is

$$F = \int_0^{\infty} \frac{1}{2} K \left( \frac{d\theta}{dz} \right)^2 - \frac{1}{2} \chi_{\perp} H^2 - \frac{1}{2} \chi_a H^2 \sin^2 \theta \, dz$$

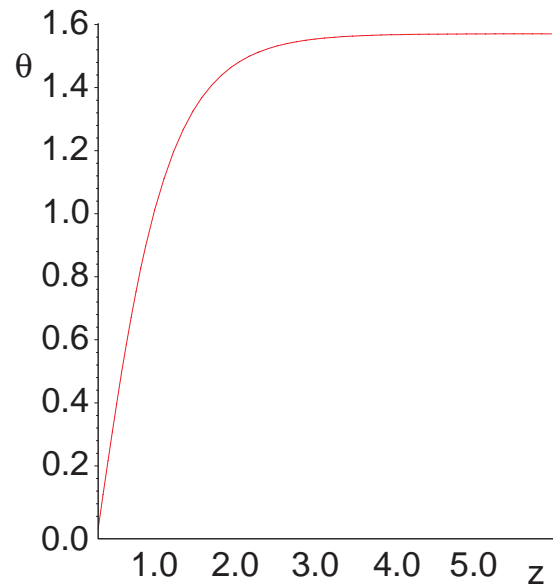
The Euler-Lagrange equation is then,

$$K \frac{d^2\theta}{dz^2} + \chi_a H^2 \sin \theta \cos \theta = 0$$

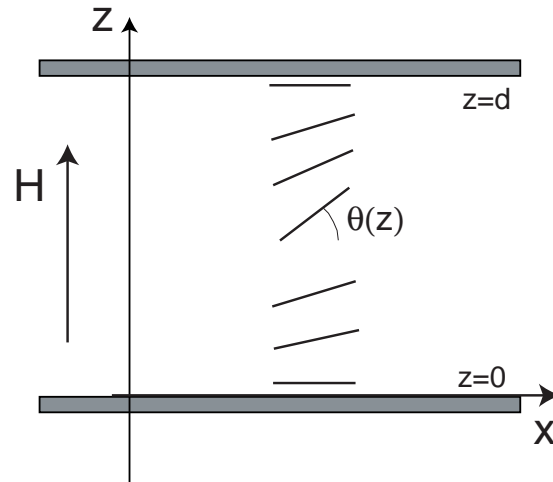
which is solved by the solution

$$\theta(z) = \pi/2 - 2 \tan^{-1} (\exp(-z/\xi))$$

where  $\xi = \frac{1}{H} \sqrt{\frac{K}{\chi_a}}$  is the magnetic coherence length.



Example: Freedericksz transition



The Euler-Lagrange equation is again,

$$K \frac{d^2\theta}{dz^2} + \chi_a H^2 \sin\theta \cos\theta = 0$$

but we must now satisfy the boundary conditions  $\theta = 0$  on  $z = 0$  and  $z = d$ .

When  $H = 0$  then this equation is solved by  $\theta(z) = Az + B$ . The boundary conditions lead to  $A = 0$  and  $B = 0$ . So  $\theta(z) \equiv 0$ , there is no distortion and the surface anchoring has determined the director within the cell.

When  $H \neq 0$  we assume that there is a small distortion  $\theta(z) \ll 1$ .

Then  $\sin \theta \approx \theta$  and  $\cos \theta \approx 1$  so that the Euler-Lagrange equation becomes

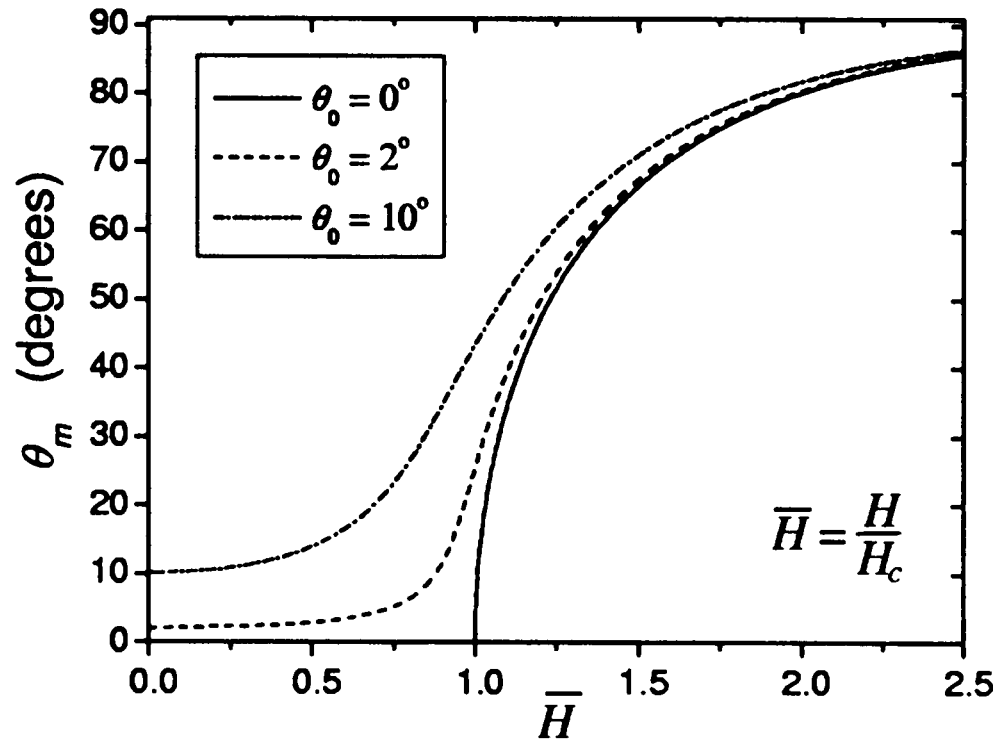
$$K \frac{d^2 \theta}{dz^2} + \chi_a H^2 \theta = 0$$

which is solved by  $\theta(z) = A \sin(z/\xi) + B \cos(z/\xi)$ . The boundary condition  $\theta(0) = 0$  gives  $B = 0$  and the boundary condition  $\theta(d) = 0$  gives a condition on the magnetic field,

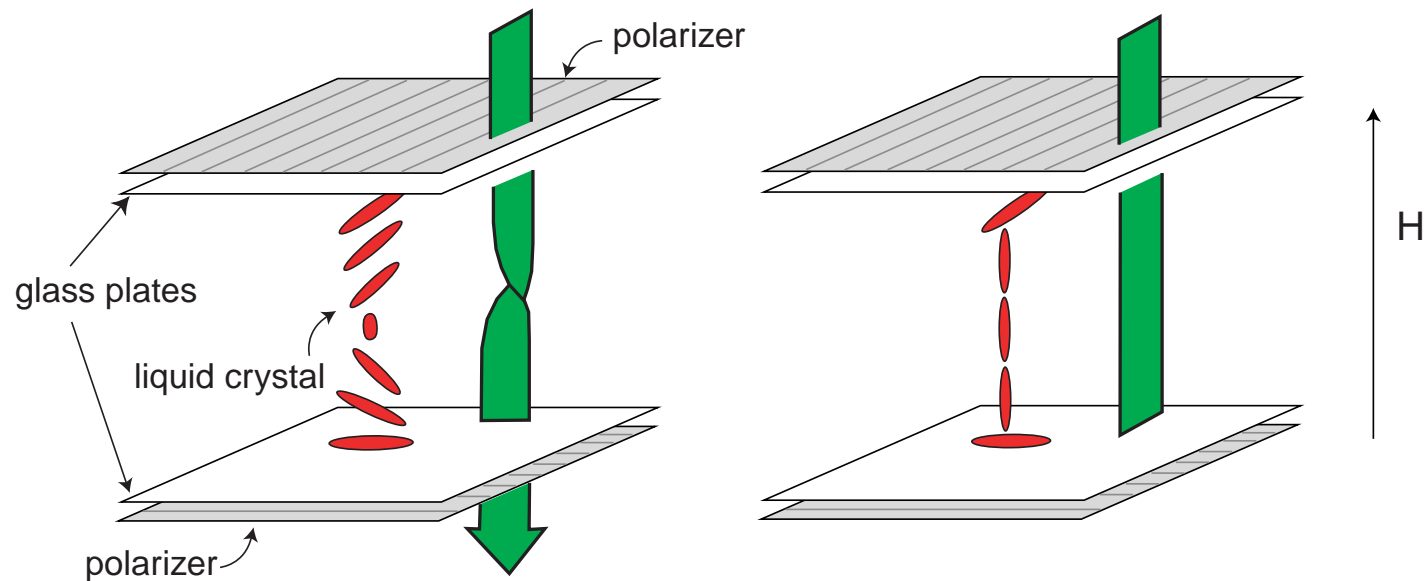
$$\begin{aligned} \xi &= \frac{d}{\pi} \\ \text{or } H &= \frac{\pi}{d} \sqrt{\frac{K}{\chi_a}} \end{aligned}$$

This critical field strength is reached when the magnetic coherence length is of the same order as the cell thickness  $d$ .

If the alignment layer induces a pretilt at the cell substrate then the distortion of the director occurs at any field strength.



## Twisted Nematic device



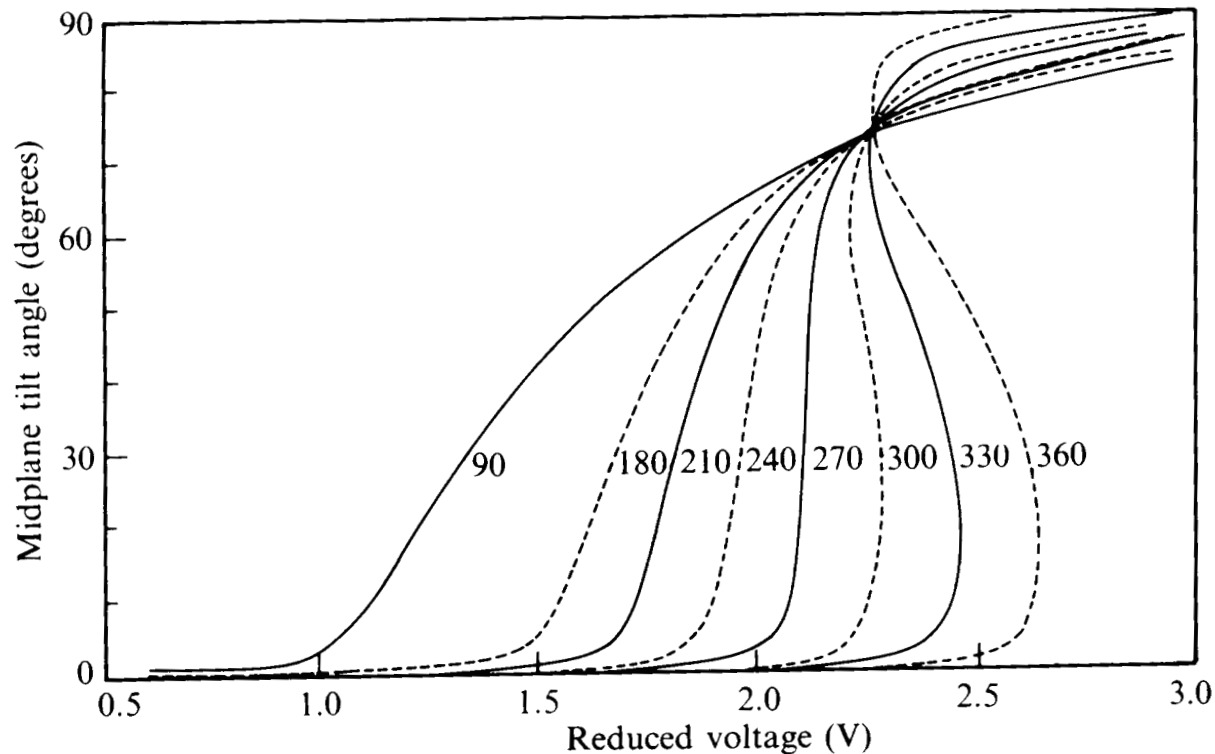
The equations are much more complicated in this case since the initial state is a twisted director structure. But the critical magnetic field strength can be found analytically to be,

$$H_c^2 = \frac{1}{\chi_a} \left( \frac{\pi}{d} \right)^2 \left( K_1 + \frac{1}{4}(K_3 - 2K_2) \right)$$

## Super-Twist Nematic device

When the amount of twist in the cell is increased the form of the transition from unswitched to switched changes significantly.

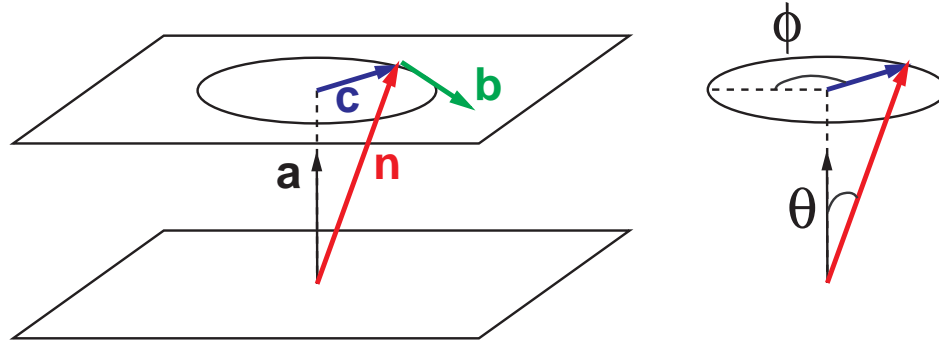
In fact, when there is  $270^\circ$  of twist the transition is almost first order and the director in the centre of the cell changes suddenly from being aligned in the plane of the substrates to aligned with the field.



## Elasticity in smectics

Because the smectic phase is more ordered than the nematic, there are less symmetry constraints on the system. This means the elastic energy is more complicated and depends on gradients of the director  $\mathbf{n}$  and the layer normal  $\mathbf{a}$ .

In fact it is easier to specify the energy in terms of the layer normal  $\mathbf{a}$ , the projection of the director onto the smectic plane  $\mathbf{c}$  and the vector perpendicular to them  $\mathbf{b} = \mathbf{a} \times \mathbf{c}$ ,



The elastic energy is,

$$\begin{aligned}
 w_d = & \frac{1}{2}K_1(\nabla \cdot \mathbf{a})^2 + \frac{1}{2}K_2(\nabla \cdot \mathbf{c})^2 + \frac{1}{2}K_3(\mathbf{a} \cdot \nabla \times \mathbf{c})^2 + \frac{1}{2}K_4(\mathbf{c} \cdot \nabla \times \mathbf{c})^2 \\
 & + \frac{1}{2}K_5(\mathbf{b} \cdot \nabla \times \mathbf{c})^2 + K_6(\nabla \cdot \mathbf{a})(\mathbf{b} \cdot \nabla \times \mathbf{c}) + K_7(\mathbf{a} \cdot \nabla \times \mathbf{c})(\mathbf{c} \cdot \nabla \times \mathbf{c}) \\
 & + K_8(\nabla \cdot \mathbf{c})(\mathbf{b} \cdot \nabla \times \mathbf{c}) + K_9(\nabla \cdot \mathbf{a})(\nabla \cdot \mathbf{c})
 \end{aligned}$$

## Flow in liquid crystals

In Newtonian fluids the stress on a area of fluid is directly proportional to the rate of strain through the viscosity of the material  $\mu$ ,

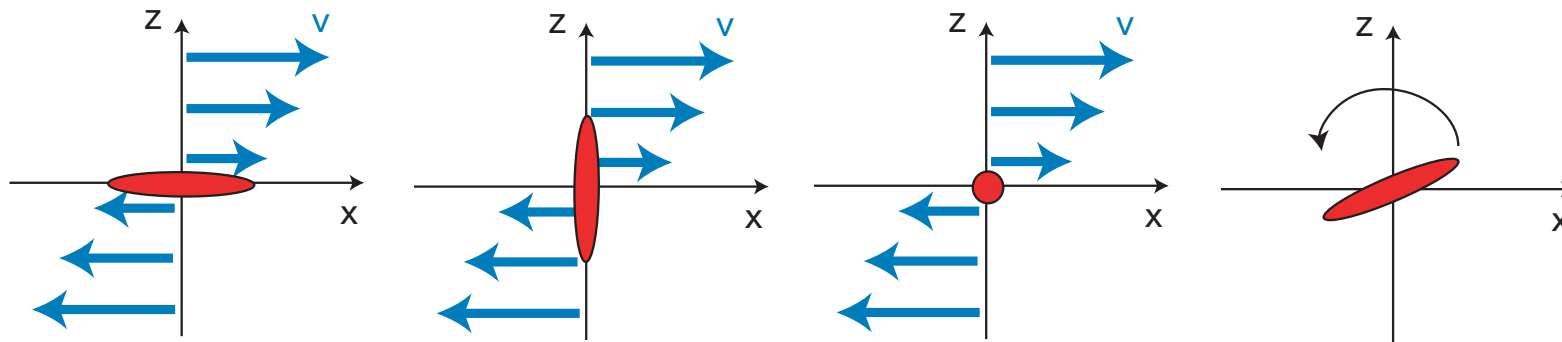
$$t_{ij} = \frac{\mu}{2} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right)$$

where  $\mathbf{v} = (v_1, v_2, v_3)$  is the velocity of the fluid at the point  $\mathbf{r} = (x_1, x_2, x_3) = (x, y, z)$ .

But in an anisotropic fluid such as a nematic the effective viscosity may depend on the director orientation.

In fact there are five independent viscosities in nematics.

Miesowicz first investigated the anisotropic nematic viscosities in the 1930's identifying three independent shear flow viscosities, a director rotation viscosity and a fifth viscosity which appears in the effective viscosity of a shear flow with the director at an arbitrary angle.



(a)  $\eta_1 = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6)$ , (b)  $\eta_2 = \frac{1}{2}(-\alpha_2 + \alpha_4 + \alpha_5)$ , (c)  $\eta_3 = \frac{1}{2}\alpha_4$ , (d)  $\gamma_1 = \alpha_3 - \alpha_2$

For both nematics and smectics a continuum theory can be constructed using the following balance laws

- continuity equation (we assume incompressibility)
- linear momentum balance (Navier-Stokes equations with a more complicated stress tensor)
- angular momentum balance (the coupling between fluid velocity and director rotation)

## Nematics: Ericksen-Leslie-(Parodi) theory

For nematics the balance equations are

mass	$v_{i,i} = 0$
linear momentum	$\rho \frac{dv_i}{dt} = \rho F_i + t_{ij,j}$
angular momentum	$G_i + g_i + s_{ij,j} = 0$

where

$\rho$  is the density,

$\mathbf{t}$  the stress tensor,

$\mathbf{F}$  is the external body force per unit mass,

$\mathbf{l} = \mathbf{n} \times \mathbf{s}$  the couple stress tensor,

$\mathbf{g}$  is the intrinsic body force and  $(e_{ijk} n_j g_k = e_{ijk} t_{kj})$

$\mathbf{K} = \mathbf{n} \times \mathbf{G}$  is the external body couple.

The stresses include contributions from pressure, elasticity and viscous forces.

the constitutive equations are

$$t_{ij} = -p\delta_{ij} - \frac{\partial w}{\partial n_{p,j}} n_{p,j} + \tilde{t}_{ij}$$

$$s_{ij} = \frac{\partial w}{\partial n_{i,j}} + \tilde{s}_{ij}$$

$$g_i = \gamma n_i - \frac{\partial w}{\partial n_i} + \tilde{g}_i$$

$$w = w(n_i, n_{i,j})$$

where  $p$  is the pressure arising from the incompressibility condition,  
 $\gamma$  is a Lagrange multiplier deriving from the  $|\mathbf{n}|^2 = 1$  condition,  
 $\tilde{\mathbf{t}}$ ,  $\tilde{\mathbf{s}}$  and  $\tilde{\mathbf{g}}$  are dynamic contributions and  
 $w$  is the elastic energy density

The dynamic contributions,  $\tilde{t}$ ,  $\tilde{s}$  and  $\tilde{g}$  are assumed to vanish in a rigid rotation so that they are functions of velocity gradients and director rotation through the rate of strain tensor  $D$  and a vector  $N$  where,

$$D_{ij} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right), \quad N_i = \frac{dn_i}{dt} - B_{ik}n_k, \quad B_{ij} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} - \frac{\partial v_j}{\partial x_i} \right)$$

The dynamic contributions must also be invariant to rotations and the transformation  $\mathbf{n} \rightarrow -\mathbf{n}$ .

the dynamic contributions are therefore,

$$\tilde{t}_{ij} = \alpha_1 n_k n_p D_{kp} n_i n_j + \alpha_2 N_i n_j + \alpha_3 N_j n_i + \alpha_4 D_{ij} + \alpha_5 D_{ik} n_k n_j + \alpha_6 D_{jk} n_k n_i$$

$$\tilde{g}_{ij} = (\alpha_2 - \alpha_3) N_i + (\alpha_5 - \alpha_6) D_{ik} n_k$$

$$\tilde{s}_{ij} = 0$$

Parodi added a restriction to the Leslie coefficients ( $\alpha_i$ ) through an Onsager relation (reversibility) so that,

$$\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$$

There are therefore five independent viscosities.

## Example: Shear flow

If  $\mathbf{v} = (v(z, t), 0, 0)$  and  $\mathbf{n} = (\cos(\theta(z, t)), 0, \sin(\theta(z, t)))$  the equations reduce to,

$$0 = -(\alpha_3 - \alpha_2) \frac{\partial \theta}{\partial t} + (K_{11} \cos^2 \theta + K_{33} \sin^2 \theta) \frac{\partial^2 \theta}{\partial z^2} + (K_{33} - K_{11}) \sin \theta \cos \theta \left( \frac{\partial \theta}{\partial z} \right)^2 - (\alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta) \frac{\partial v}{\partial z}$$

$$0 = -\rho \frac{\partial v}{\partial t} + \frac{\partial}{\partial z} \left( \left( \frac{\alpha_4}{2} + \frac{(\alpha_5 - \alpha_2)}{2} \sin^2 \theta + \frac{(\alpha_6 + \alpha_3)}{2} \cos^2 \theta + \alpha_1 \sin^2 \theta \cos^2 \theta \right) \frac{\partial v}{\partial z} + (\alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta) \frac{\partial \theta}{\partial t} \right)$$

And if we consider steady shear flow  $v = \kappa z$  with constant  $\theta$  then the equations reduce to

$$(\alpha_3 \cos^2 \theta - \alpha_2 \sin^2 \theta) = 0$$

so that  $\theta = \tan^{-1} \sqrt{\frac{\alpha_3}{\alpha_2}}$ .

The shear flow has aligned the director.

## Neglecting flow

If we use the Ericksen-Leslie equations but assume that the flow is negligible (not always a very accurate assumption) then we are left with equations for the director rotation.

Example: In the Freedericksz cell, if we set  $\mathbf{v} = \mathbf{0}$  the only equation we have left is,

$$(\alpha_3 - \alpha_2) \frac{\partial \theta}{\partial t} = K \frac{\partial^2 \theta}{\partial z^2} + \chi_a H^2 \sin \theta \cos \theta$$

Before the distortion occurs at the critical field strength we know that  $\theta(z, t) \equiv 0$ .

After the transition occurs the director will distort with  $\theta$  increasing in the middle of the cell.

If we assume  $\theta$  is at least initially small and set  $\theta(z, t) = A \exp(t/\tau) \sin(\pi z/d)$  then if we find that  $\tau$  is negative, an initial distortion will be damped down to leave the  $\theta \equiv 0$ , undistorted state.

If however we find that  $\tau$  is positive then any initial distortion will increase. The rate of increase, i.e. the switching rate, is then measured by  $\tau$ . The smaller  $\tau$  is the faster the switching occurs.

We find,

$$\frac{(\alpha_3 - \alpha_2)}{\tau} = - \left( \frac{\pi}{d} \right)^2 K + \chi_a H^2$$

so that

$$\tau = \frac{\alpha_3 - \alpha_2}{\chi_a H^2 - K \pi^2 / d^2}$$

Therefore, if  $\chi_a > 0$  and  $H^2 > \frac{K \pi^2}{\chi_a d^2}$  then  $\tau$  is positive and switching will occur.

If  $\chi_a < 0$  then  $\tau$  is always negative so the system will always revert to the  $\theta \equiv 0$  state.

## Dynamic theory of smectics

An equivalent theory of flow in smectics does exist although it is a lot more complicated. Only simple cell geometries have been modelled.

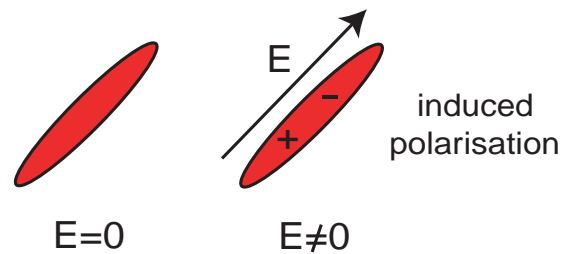
The main problem with this theory, called the Leslie-Stewart-Nakagawa theory, is that it is much harder to understand the various viscosities. This is mainly due to the fact that there are 20 of them!

$$\begin{aligned}
 \tilde{t}_{ij} = & \mu_0 D_{ij} + \mu_1 a_p D_p^a a_i a_j + \mu_2 (D_i^a a_j + D_j^a a_i) + \mu_3 c_p D_p^c c_i c_j + \mu_4 (D_i^c c_j + D_j^c c_i) \\
 & + \mu_5 c_p D_p^a (a_i c_j + a_j c_i) + \lambda_1 (A_i a_j + A_j a_i) + \lambda_2 (C_i c_j + C_j c_i) \\
 & + \lambda_3 c_p A_p (a_i c_j + a_j c_i) + \kappa_1 (D_i^a c_j + D_j^a c_i + D_i^c a_j + D_j^c a_i) \\
 & + \kappa_2 [a_p D_p^a (a_i c_j + a_j c_i) + 2a_p D_p^c a_i a_j] + \kappa_3 [c_p D_p^c (a_i c_j + a_j c_i) + 2a_p D_p^c c_i c_j] \\
 & + \tau_1 (C_i a_j + C_j a_i) + \tau_2 (A_i c_j + A_j c_i) + 2\tau_3 c_p A_p a_i a_j + 2\tau_4 c_p A_p c_i c_j \\
 & + \lambda_1 (D_j^a a_i - D_i^a a_j) + \lambda_2 (D_j^c c_i - D_i^c c_j) + \lambda_3 c_p D_p^a (a_i c_j - a_j c_i) \\
 & + \lambda_4 (A_j a_i - A_i a_j) + \lambda_5 (C_j c_i - C_i c_j) + \lambda_6 c_p A_p (a_i c_j - a_j c_i) \\
 & + \tau_1 (D_j^a c_i - D_i^a c_j) + \tau_2 (D_j^c a_i - D_i^c a_j) + \tau_3 a_p D_p^a (a_i c_j - a_j c_i) \\
 & + \tau_4 c_p D_p^c (a_i c_j - a_j c_i) + \tau_5 (A_j c_i - A_i c_j + C_j a_i - C_i a_j)
 \end{aligned}$$

## Lecture 2: Modelling Electric Fields

- Dielectric effect: Induced polarisation
- Solving Maxwell's equations in the cell
- Spontaneous polarisation
- Alignment layers
- Modelling ferroelectric LC devices

The electron distribution in a liquid crystal molecule will distort if placed in an external electric field.



This distortion will induce a small molecular dipole which in turn influences the external electric field.

There is feedback between the externally applied field and the molecular field.

In the magnetic field case this feedback was very weak because the susceptibility to such an effect was extremely weak (remember  $\chi_{||}$  and  $\chi_{\perp}$  were about  $10^{-6}$ ).

However, the dielectric susceptibilities are much stronger, typically  $\chi_{e||}$  and  $\chi_{e\perp}$  are of order 1. So the liquid crystal molecules will significantly influence the electric field and it cannot be taken to be constant as in the magnetic case.

The induced polarisation  $\mathbf{P}$  (equivalent to the magnetisation  $\mathbf{M}$ ) is related to the applied electric field through the susceptibilities,

- If  $\mathbf{n}$  is parallel to  $\mathbf{E}$  the polarisation is  $\mathbf{P} = \chi_{e\parallel} \mathbf{E}$
- If  $\mathbf{n}$  is perpendicular to  $\mathbf{E}$  the polarisation is  $\mathbf{P} = \chi_{e\perp} \mathbf{E}$

or in tensor form in the molecular coordinate system,

$$\mathbf{P} = \chi_e \mathbf{E}, \quad \chi_e = \begin{pmatrix} \chi_{e\parallel} & 0 & 0 \\ 0 & \chi_{e\perp} & 0 \\ 0 & 0 & \chi_{e\perp} \end{pmatrix}$$

the 11 term in the tensor corresponds to the long axis of the molecule.

The electric displacement  $\mathbf{D}$  (equivalent to the magnetic induction  $\mathbf{B}$ ) is then

$$\mathbf{D} = \epsilon_0 (\mathbf{E} + \mathbf{P})$$

so,

$$\mathbf{D} = \epsilon_0 ((\mathbf{I} + \chi_e) \mathbf{E}) = \epsilon_0 \epsilon^{mol} \mathbf{E}$$

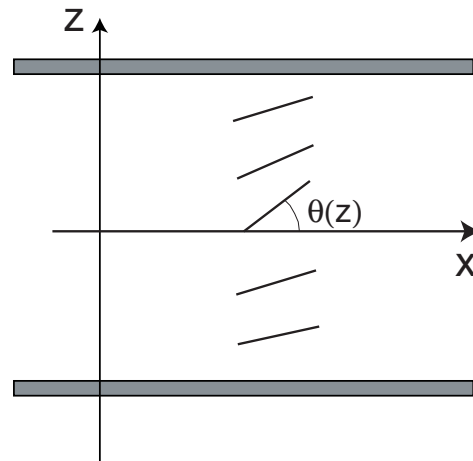
where  $\epsilon^{mol} = \mathbf{I} + \chi_e$  is called the dielectric tensor in the molecular frame of reference.

In a general frame of reference (i.e. the laboratory frame) the dielectric tensor must be transformed.

If the rotation matrix  $R$  takes the molecular frame of reference to the laboratory frame of reference then the dielectric tensor in the laboratory frame is

$$\epsilon^{lab} = \mathbf{R}\epsilon^{mol}\mathbf{R}^T$$

Example: simple nematic cell



In this case the laboratory frame and the molecular frame differ by a rotation by  $\theta$  around the  $y$  axis.

This rotation has the form

$$\mathbf{R} = \begin{pmatrix} \cos \theta & 0 & -\sin \theta \\ 0 & 1 & 0 \\ \sin \theta & 0 & \cos \theta \end{pmatrix}$$

and so

$$\begin{aligned} \epsilon^{lab} &= \mathbf{R} \epsilon^{mol} \mathbf{R}^T \\ &= \begin{pmatrix} \cos \theta & 0 & -\sin \theta \\ 0 & 1 & 0 \\ \sin \theta & 0 & \cos \theta \end{pmatrix} \begin{pmatrix} \epsilon_{||} & 0 & 0 \\ 0 & \epsilon_{\perp} & 0 \\ 0 & 0 & \epsilon_{\perp} \end{pmatrix} \begin{pmatrix} \cos \theta & 0 & \sin \theta \\ 0 & 1 & 0 \\ -\sin \theta & 0 & \cos \theta \end{pmatrix} \\ &= \begin{pmatrix} \epsilon_{||} \cos^2 \theta + \epsilon_{\perp} \sin^2 \theta & 0 & (\epsilon_{||} - \epsilon_{\perp}) \sin \theta \cos \theta \\ 0 & \epsilon_{\perp} & 0 \\ (\epsilon_{||} - \epsilon_{\perp}) \sin \theta \cos \theta & 0 & \epsilon_{\perp} \cos^2 \theta + \epsilon_{||} \sin^2 \theta \end{pmatrix} \end{aligned}$$

From now on we'll always use the laboratory frame of reference so we drop the <sup>lab</sup> superscript.

In the laboratory frame we then have  $\mathbf{D} = \epsilon_0 \epsilon \mathbf{E}$ .

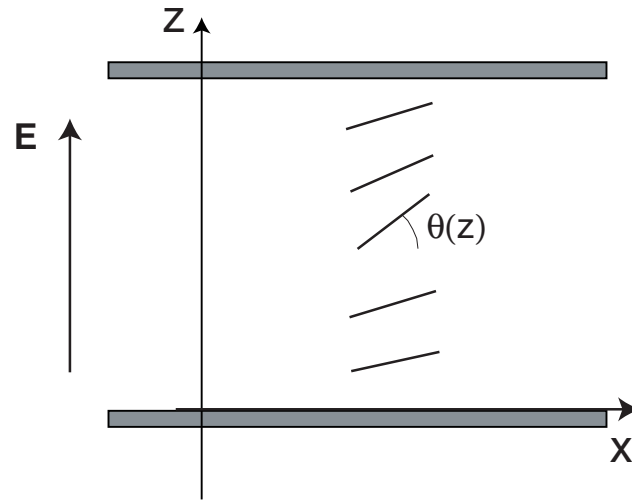
The electrostatic energy is (similar to the magnetic case)

$$\begin{aligned} w_e &= -\frac{1}{2} \mathbf{D} \cdot \mathbf{E} \\ &= -\frac{1}{2} \epsilon_0 \epsilon \mathbf{E} \cdot \mathbf{E} \end{aligned}$$

But we don't know  $\mathbf{E}$ ! This is calculated using Maxwell's equations.

There are two approaches to solving for the director structure and the electric field simultaneously and I will demonstrate both with the aid of the simple example we have used throughout.

Example: The simple 1-dimensional nematic cell.



We can assume that  $\theta$ ,  $\mathbf{D} = (D_1, D_2, D_3)$  and  $\mathbf{E} = (E_1, E_2, E_3)$  are functions of  $z$  only.

## Method 1

Maxwell's equations (with no free charge) which are,

$$\nabla \times \mathbf{E} = 0$$

$$\nabla \cdot \mathbf{D} = 0$$

become,

$$\frac{dD_3}{dz} = \frac{dE_1}{dz} = \frac{dE_2}{dz} = 0$$

There is no voltage applied in the  $x$  and  $y$  directions so we can assume that  $E_1 = E_2 = 0$ . We also now know that  $D_3$  does not depend on  $z$

The relationship between  $\mathbf{D}$  and  $\mathbf{E}$  gives

$$\begin{aligned}\mathbf{D} &= \epsilon_0 \epsilon \mathbf{E} \\ D_3 &= \epsilon_0 \epsilon_{33} E_3\end{aligned}$$

where  $\epsilon_{33} = \epsilon_{\perp} \cos^2 \theta + \epsilon_{\parallel} \sin^2 \theta$

So we have  $\mathbf{E} = (0, 0, E_3)$  in terms of the, as yet, unknown constant  $D_3$ .

Integrating the electric field gives

$$\begin{aligned}\int_0^d E_3 dz &= \int_0^d \frac{D_3}{\epsilon_0 \epsilon_{33}} dz \\ &= D_3 \int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz\end{aligned}$$

But the the electric field is defined in terms of the gradient of the electric potential  $\mathbf{E} = -\nabla U$  which gives  $E_3 = -\frac{dU}{dz}$  so

$$\int_0^d E_3 dz = \int_0^d -\frac{dU}{dz} dz = -U(d) + U(0)$$

If the voltage is applied at  $z = 0$  and the electrode at  $z = d$  is grounded then  $U(d) = 0$ ,  $U(0) = V$ .

so

$$V = D_3 \int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz$$

or equivalently,

$$D_3 = \frac{V}{\int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz}$$

and then the electric field is,

$$E_3 = \frac{1}{\epsilon_0 \epsilon_{33}} \frac{V}{\int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz}$$

which can be used to find the electric energy.

Minimisation of this energy is not straightforward but after a length calculation we find a similar equation to the magnetic case, The Euler-Lagrange equation is again,

$$K \frac{d^2 \theta}{dz^2} + \epsilon_0 \epsilon_a E_3^2 \sin \theta \cos \theta = 0$$

but  $E_3$  must be calculated simultaneously from

$$E_3 = \frac{1}{\epsilon_0 \epsilon_{33}} \frac{V}{\int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz}$$

where  $\epsilon_{33} = \epsilon_{\perp} \cos^2 \theta + \epsilon_{\parallel} \sin^2 \theta$ .

An analysis of this equation gives the critical voltage as,

$$V_c = \pi \sqrt{\frac{K}{\epsilon_0 \epsilon_a}}$$

## Method 2

In this method we will produce equations for both  $\theta(z)$  and the electric potential  $U(z)$ . It looks simpler but if you want to consider dynamics it is sometimes easier to solve using Method 1.

The Maxwell equation  $\nabla \times \mathbf{E} = 0$  implies that there exists a potential function  $U$  such that  $\mathbf{E} = -\nabla U$  which we have seen means that  $E_3 = -\frac{dU}{dz}$ .

The electric energy is then

$$\begin{aligned} w_e &= -\frac{1}{2} \mathbf{D} \cdot \mathbf{E} \\ &= -\frac{1}{2} \epsilon_0 \epsilon \mathbf{E} \cdot \mathbf{E} = -\frac{1}{2} \epsilon_0 \epsilon_{33} \left( \frac{dU}{dz} \right)^2 \end{aligned}$$

which when added to the elastic energy we have the total energy,

$$F = \int_0^\infty \frac{1}{2} K \left( \frac{d\theta}{dz} \right)^2 - \frac{1}{2} \epsilon_0 \epsilon_{33} \left( \frac{dU}{dz} \right)^2 dz$$

The Euler-Lagrange equations for  $\theta(z)$  and  $U(z)$  are then,

$$K \frac{d^2\theta}{dz} + \frac{1}{2} \epsilon_0 \frac{d\epsilon_{33}}{d\theta} \left( \frac{dU}{dz} \right)^2 = 0$$
$$\frac{d}{dz} \left( \frac{1}{2} \epsilon_0 \epsilon_{33} \left( \frac{dU}{dz} \right) \right) = 0$$

These equations must then be solved simultaneously with the boundary conditions  $\theta(d) = 0$ ,  $\theta(0) = 0$  and  $U(d) = V$ ,  $U(0) = 0$ .

Remember  $\epsilon_{33} = \epsilon_{\perp} \cos^2 \theta + \epsilon_{\parallel} \sin^2 \theta$  and  $\theta$  is a function of  $z$ .

## Spontaneous polarisation

In chiral smectic C materials there are two major differences to the nematic case when calculating the electric field.

The first is that the molecular distribution is biaxial. This means that the dielectric tensor in the molecular frame of reference has three distinct values on the diagonal,

$$\epsilon^{mol} = \begin{pmatrix} \epsilon_1 & 0 & 0 \\ 0 & \epsilon_2 & 0 \\ 0 & 0 & \epsilon_3 \end{pmatrix}$$

The second, and very important, difference is the fact that there exists a permanent macroscopic polarisation even when there is no applied voltage.

This polarisation is in the direction perpendicular to  $\mathbf{n}$  and  $\mathbf{a}$ .

The total polarisation at a point in the cell is therefore the sum of the dielectrically induced polarisation and the spontaneous polarisation.

$$\mathbf{P}_{total} = \mathbf{P}_d + \mathbf{P}_s = \epsilon_0 \epsilon \mathbf{E} + \mathbf{P}_s$$

The calculation of the electric field will change and we find that,

$$E_3 = \frac{1}{\epsilon_0 \epsilon_{33}} \frac{V + \int_0^d \frac{P_{s3}}{\epsilon_0 \epsilon_{33}} dz}{\int_0^d \frac{1}{\epsilon_0 \epsilon_{33}} dz}$$

## Alignment layers

In real devices the electrode is rarely in contact with the liquid crystal material.

There is usually an alignment layer between the electrode and LC which will alter the effective electric field that the liquid crystal molecules experience.

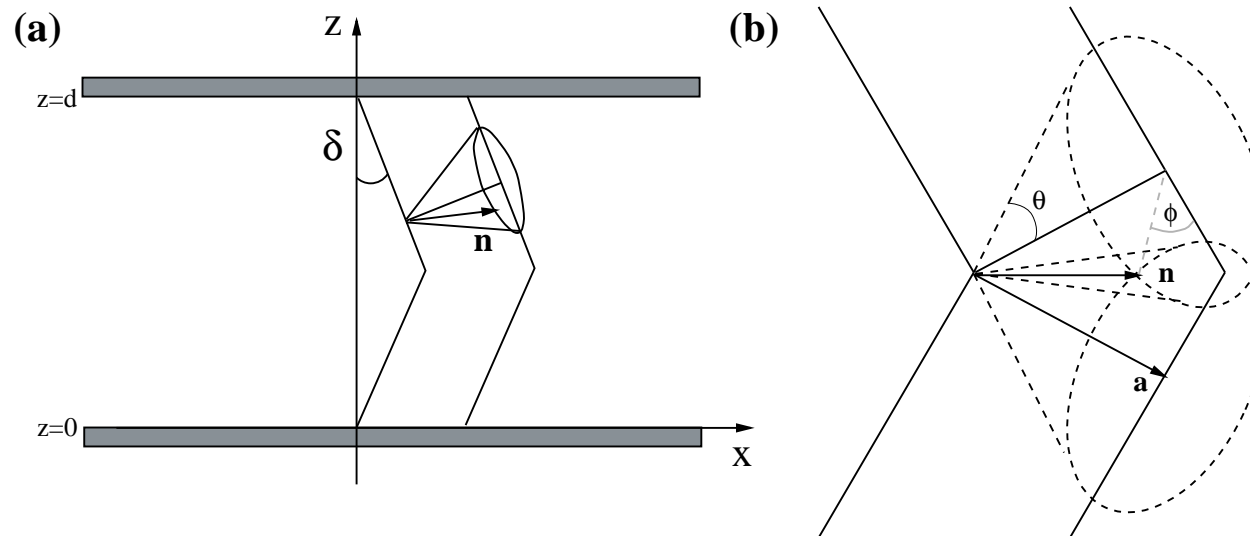
In a full model of a device the electric field must be calculated within the alignment layer and the field  $\mathbf{E}$  must be matched at the boundary between the alignment layer and LC.

However, solving Maxwell's equations in the alignment layer is a relatively straightforward calculation since the alignment layer orientation is fixed unlike the LC where the director orientation changes.

## Modelling Ferroelectric LC devices

In a typical ferroelectric liquid crystal (FLC) device the layers form a chevron structure.

At the chevron interface the director can take one of two orientations. There is an energy barrier between these two orientations so the cell is **bistable**



The smectic cone angle is fixed to be  $\theta$  and the smectic layer tilt angle is  $\delta$ . The voltage  $V$  is applied across the cell which is of thickness  $d$ .

A simple theory can be constructed which considers only the director at the chevron interface.

The director at the interface is assumed to lie on a cone of angle  $\theta$  and the azimuthal angle around the cone is denoted by  $\phi$

The interface torque on the director due to the two stable positions is,

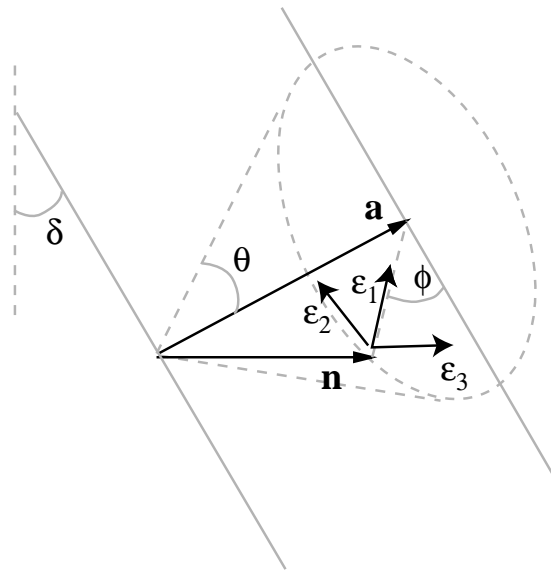
$$Q_k = B (\sin(2\phi) - 2 \cos \phi_{chev} \sin \phi)$$

where  $B$  is an effective elastic constant and  $\pm\phi_{chev}$  are two stable director positions around the cone determined by the chevron configuration so that  $\cos \phi_{chev} = \tan \delta / \tan \theta$

The dielectric torque, which derives from the biaxial dielectric tensor, is

$$Q_e = \frac{\epsilon_0(\partial\epsilon - \Delta\epsilon \sin^2 \theta)V^2}{2d^2} \cos^2 \delta \sin(2\phi) + \frac{\epsilon_0 \Delta\epsilon \sin(2\theta)V^2}{2d^2} \sin \delta \cos \delta \sin \phi$$

The dielectric anisotropy  $\Delta\epsilon = \epsilon_3 - \epsilon_1$  and the dielectric biaxiality is  $\partial\epsilon = \epsilon_2 - \epsilon_1$ .



The polarisation torque is

$$Q_p = \frac{P_s V}{d} \cos \delta \cos \phi$$

where  $P_s$  is the spontaneous polarisation.

The governing equation for the dynamics is then

$$\eta_c \frac{d\phi}{dt} = Q_k + Q_e + Q_p$$

By applying a voltage waveform  $V(t)$  it will be possible to switch the director at the chevron between the two stable positions  $+\phi_0$  and  $-\phi_0$  through the polarisation torque.

Effects due to the dielectric biaxiality of the smectic C phase can be investigated using this model.