MALVERN PHYSICS SERIES

General Series Editor: **Professor E R Pike** FRS

NONLINEAR PHENOMENA AND CHAOS

Edited by **Sarben Sarkar**
Royal Signals and Radar Establishment, Malvern

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PREFACE

This volume contains most of the invited talks given at the Malvern Seminar on Dynamical Systems held at RSRE, Malvern, on 23–25 April 1985.

The topics presented deal with fluids, chemical reactions, nonlinear and quantum optics, and theories of Hamiltonian and dissipative systems.

I would like to thank Professor E R Pike for his encouragement and support of the idea of the Seminar, and for editing these proceedings as the first volume in the Malvern Physics Series, and also the Director and Board of Management of RSRE for funding it. I am grateful to Drs D S Broomhead and G P King for their invaluable help during the planning stages, and above all to Mrs D Costanzo and Mrs M F Allso of the Symposium Secretariat for their superb organisation of the meeting.

Sarben Sarkar
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April 1985

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CONTENTS

FLUCTUATIONS, OSCILLATIONS AND INSTABILITIES IN LIQUID CRYSTAL DEVICES

M G CLARK

1. INTRODUCTION

Liquid crystal devices (LCD) have received considerable attention in
recent years, primarily because of their application as flat panel
displays. However, it has also been recognised that they provide excellent
examples of some of the most interesting phenomena displayed by nonlinear
dynamical systems. Indeed, these phenomena are important for the
understanding and development of LCD for applications. Electrohydro-
dynamically induced turbulence (called 'dynamic scattering') has been used
as a display effect (Goodman 1975). Another particularly unusual motional
instability is a problem in large area LCD shutters fast switched by the
dual-frequency technique (Raynes and Shanks 1974) unless the drive
conditions needed to avoid this are properly understood (Clark and Shanks
1982).

In the next section we introduce briefly some concepts from liquid
crystal physics which are central to the subject matter of this chapter.
The transitions which may occur in nematic liquid crystal devices are
discussed in §3, with emphasis on the concomitant fluctuations, and on
the potential for bistability or oscillations introduced by feedback. In
§4 the dynamical equations of nematic LCD are discussed in more detail.
Finally, in §5 we discuss the transition to turbulence in LCD, with
emphasis on the unusual effects which may be observed in dual-frequency
fast switching of LCD shutters.

Even the relatively well understood nematic LCD would hardly be sought
out by theoreticians as simple examples of dynamical systems. The
anisotropy and multiple hydrodynamic variables lead to a multi-dimensional
phase space, while the dynamical equations in general pose formidable
problems of solution. On the other hand, liquid crystals, by their nature,
show phenomena unknown in isotropic materials. Further, the characteristic
response times and the construction of LCD are such that the phenomena

which occur in them are easy to demonstrate in laboratory or lecture room, often by use of a standard overhead projector. At appropriate points in this chapter we describe experimental demonstrations which provide convenient illustrations of the phenomena discussed.

2. CONTINUUM THEORY

The liquid crystal phases of interest for devices are 'thermotropic', i.e. they appear as a function of temperature between the crystal and isotropic liquid phases in pure compounds or mixtures of constant composition. The phase of primary interest here is the nematic, although the cholesteric and smectic A phases will also be mentioned.

The molecules forming these phases are elongated; thus we associate with each molecule a 'long axis' (the rigorous definition of which is not without difficulty (Clark 1985)). In the nematic phase, whereas the molecular centres of mass are randomly arranged, the long axes show partial orientational ordering. The average preferred direction of the molecular long axes is called the 'director'. In continuum terms, a nematic is a fluid with local uniaxial symmetry; the director $n(r,t)$ is a unit vector parallel to this symmetry axis. Clearly the director and the molecular long axis are distinct concepts, although the literature is unfortunately often careless or misleading on this point. Only the director is a continuum macroscopic property, and only the director $n(r,t)$ (and of course the fluid velocity $v(r,t)$) need be considered to describe the operation of a nematic LCD.

The cholesteric phase is closely related, being a chiral nematic in which the local order remains nematic-like but the director adopts a helical twisted configuration in the absence of external fields or forces. In the smectic A phase the orientational ordering is again similar to a nematic, but the distribution of molecular centres of mass is no longer random, showing a mass density wave

$$\rho(z) = \rho_o[1 + \sqrt{\tfrac{1}{2}}|\psi|\cos(q_s z - \phi)] \tag{2.1}$$

where the wavevector q_s is parallel to the director n and the complex order parameter $\psi = |\psi|\exp(i\phi)$ characterises the amplitude and phase of the mass density wave.

Figure 1 illustrates the fluid structures of the three phases schematically.
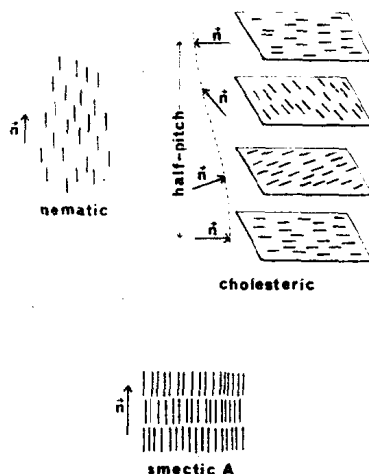
Figure 1. Schematic representation of the molecular arrangements in the nematic, cholesteric and smectic A liquid crystal phases.

The uniaxial symmetry of the nematic phase is reflected in the symmetries of property tensors. For example, the permittivity $\varepsilon(r,\omega)$ may be written

$$\varepsilon_{ij} = \frac{1}{3}(\varepsilon_{\parallel} + 2\varepsilon_{\perp})\delta_{ij} + (\varepsilon_{\parallel} - \varepsilon_{\perp})(n_i n_j - \frac{1}{3}\delta_{ij}) \qquad (2.2)$$

$$= \varepsilon_{\perp}\delta_{ij} + (\varepsilon_{\parallel} - \varepsilon_{\perp})n_i n_j \qquad (2.3)$$

where $\parallel$ and $\perp$ denote the principal components parallel and perpendicular to n, and the quantities

$$\varepsilon_{\parallel} - \varepsilon_{\perp} = \Delta\varepsilon \qquad (2.4)$$

$$\frac{1}{3}(\varepsilon_{\parallel} + 2\varepsilon_{\perp}) = \bar{\varepsilon} \qquad (2.5)$$

are the anisotropy and spherical mean, respectively. In general the 'static' permittivity ($\omega \to 0$) contains contributions from both the molecular polarisability tensor and the molecular dipole. According to the magnitude of the dipole and its orientation relative to the

molecular 'long axis', Δε may be positive or negative.

The frequency dependence of ε in a nematic is different for the
/ and ⊥ components. The lowest frequency relaxation occurs in $\varepsilon_/$, and may
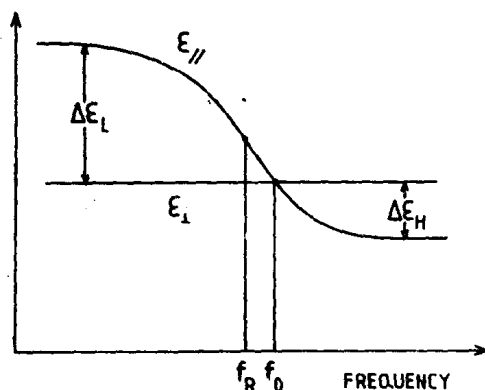in some materials be at kilohertz frequencies or lower. Thus if the

Figure 2. Definitions of the dielectric parameters of a
dual-frequency nematic material.

relative values of $\varepsilon_/$ and $\varepsilon_\perp$ are correctly arranged a material may have
Δε positive at low frequencies, and negative above some 'cross-over'
frequency' f_0 (figure 2) which lies well within the range convenient for
electronics. Materials of this kind are termed 'dual-frequency' materials;
Table I lists the parameters of such a nematic material.

This lowest frequency relaxation in nematic liquids is a good fit to
the Debye form

$$\varepsilon_/(f) = \varepsilon_/^H + \varepsilon_/^L - \varepsilon_/^H)/[1 + (f/f_R)^2] \tag{2.6}$$

where L = low and H = high frequency, and f_R is the 'relaxation frequency'
(figure 2). It is noteworthy that this relaxation in an anisotropic
liquid is a better example of the Debye form than the isotropic liquids
which the model was intended to describe.

3. FREEDERICKSZ TRANSITIONS

One consequence of the dielectric anisotropy is that the director may be
aligned by application of an external electric field. If we take an

Table 1. Material parameters of a state-of-the-art dual-frequency nematic suitable for commercial applications (McDonnell and Smith 1982).

Phase range	
solid melts	$< - 25\ ^\circ C$
(usually supercools, $T_g = - 65\ ^\circ C$)	
nematic to isotropic	$101\ ^\circ C$
Dielectric properties at 20 $^\circ$C	
$\Delta\epsilon$ at 100 Hz	$+ 2.95$
$\Delta\epsilon$ at 40 kHz	$- 1.8$
$\epsilon_\perp$	5.25
cross-over frequency f_0	5.1 kHz
Birefringence (20 $^\circ$C) Δn^D	0.12
Viscosity (20 $^\circ$C)	98 cP

electric displacement $D(z)$ and assume that the conductivity of the material is negligible (a more general case is discussed elsewhere (Clark 1985, Thurston 1984)):

$$\mathrm{div}\mathbf{D} = 0 = \partial D_z/\partial z \qquad (3.1)$$

which implies that D_z has no z dependence. Hence the electrostatic energy density

$$u_E = \tfrac{1}{2} E_z D_z = \tfrac{1}{2} D_z^2 /\epsilon_o \epsilon(\theta) \qquad (3.2)$$

where

$$\epsilon(\theta) = \epsilon_{/\!/} \cos^2\theta + \epsilon_\perp \sin^2\theta \qquad (3.3)$$

θ being the angle between $\mathbf{D}$ and $\mathbf{n}$. It follows that the director will tend to align parallel to the field if $\Delta\epsilon > 0$ and perpendicular to the field if $\Delta\epsilon < 0$.

Any distortion of the nematic director from uniform alignment is opposed by the curvature elasticity associated with the director. The elastic free energy density associated with a distortion of $\mathbf{n}(\mathbf{r})$ from the state of uniform alignment $\mathbf{n}(\mathbf{r}) = \mathbf{n}_0$ is given by (de Gennes 1974, Ch 3)

$$u_d = \tfrac{1}{2}\{K_{11}(\text{div}\mathbf{n})^2 + K_{22}(\mathbf{n}.\text{curl}\mathbf{n})^2 + K_{33}(\mathbf{n}\times\text{curl}\mathbf{n})^2$$

$$+ K_{24}\text{div}[(\mathbf{n}.\text{grad})\mathbf{n} - \mathbf{n}\text{div}\mathbf{n}]\} \tag{3.4}$$

where K_{11}, K_{22} and K_{33} are elastic constants corresponding respectively to splay, twist, and bend bulk distortions of the director, and it follows from the divergence theorem that K_{24} contributes only to surface energies.

The competition between an electric field seeking to realign the director and elastic forces resisting that realignment is exploited in liquid crystal devices such as that shown in figure 3. A thin film of
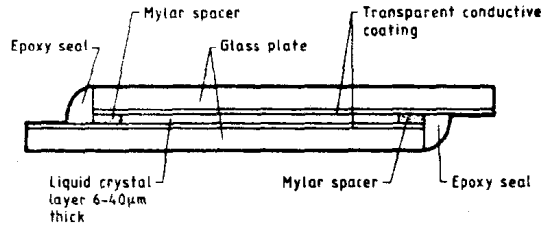

Figure 3. Construction of a typical liquid crystal cell.

liquid crystal, typically 5 - 10 μm thick but sometimes up to 50 μm, is confined between two glass plates. The inner surfaces of the plates bear transparent electrodes (indium tin oxide) which are used to apply an electric field across the film. In addition they bear surface alignment coatings. Various kinds of coating are used, their essential function being to fix the direction of **n** at the cell walls (Castellano 1984). If the director is pinned parallel to the cell wall the alignment is called 'homogeneous', if **n** makes a slight angle with the wall the alignment is 'tilted homogeneous', and if **n** is pinned perpendicular to the wall the alignment is 'homeotropic'.

The simplest kind of LCD is a parallel aligned cell, in which the direction of homogeneous alignment is the same on both walls. In the absence of applied voltage the director then assumes a uniform parallel alignment throughout the cell (figure 4(a)). Now let an electric field be applied across the cell, and consider a perturbation in which the director becomes tilted at an angle α to the plane of the cell, given by
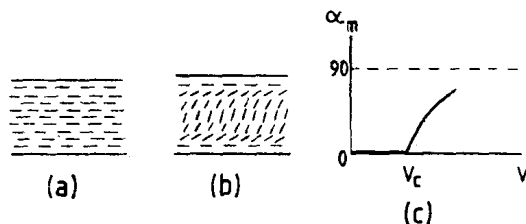
Figure 4. Freedericksz transition in a parallel aligned
nematic LCD; (a) director configuration when $V < V_c$,
(b) director configuration when $V > V_c$, (c) plot of tilt
angle at centre of cell, α_m, against applied RMS voltage V.

$$\alpha = \alpha_m \sin(\pi z/d) \tag{3.5}$$

where z is measured perpendicular to the plane of the cell, d is the
thickness of the nematic film, and α_m is the tilt at the centre of the
cell. The change in free energy per unit area of cell is then

$$d^{-1} \int_o^d \delta(u_d + u_E) dz = \tfrac{1}{4} K_{11}(\pi/d)^2 \alpha_m^2 - \tfrac{1}{4}\epsilon_o \Delta\epsilon (D_z/\epsilon_o \epsilon_\perp)^2 \alpha_m^2 + O(\alpha_m^4). \tag{3.6}$$

Evidently the free energy per unit area is <u>decreased</u> by distortion if

$$(D_z^2 \Delta\epsilon/\epsilon_o \epsilon_\perp^2) > (\pi^2 K_{11}/d^2) \tag{3.7}$$

i.e. the director configuration distorts as shown in figure 4(b) above a
critical voltage

$$V_c = \pi(K_{11}/\epsilon_o \Delta\epsilon)^{\frac{1}{2}}. \tag{3.8}$$

The transition has all the features of a second-order phase transition of
the 'instability' type (de Gennes 1975); α_m is an order parameter of the
transition (figure 4(c)); a generalised susceptibility which diverges at
the critical voltage V_c may be defined (Forster 1975).

It is convenient to demonstrate these features using the 'twisted
nematic' type of LCD employed in most practical applications. In this
device the directions of homogeneous alignment on the two cell walls are
at right angles. Thus, in the absence of applied voltage, the director
adopts a 90° twist (figure 5(a)). This twisted birefringent structure
will rotate the plane of plane polarised light by 90°, causing light to
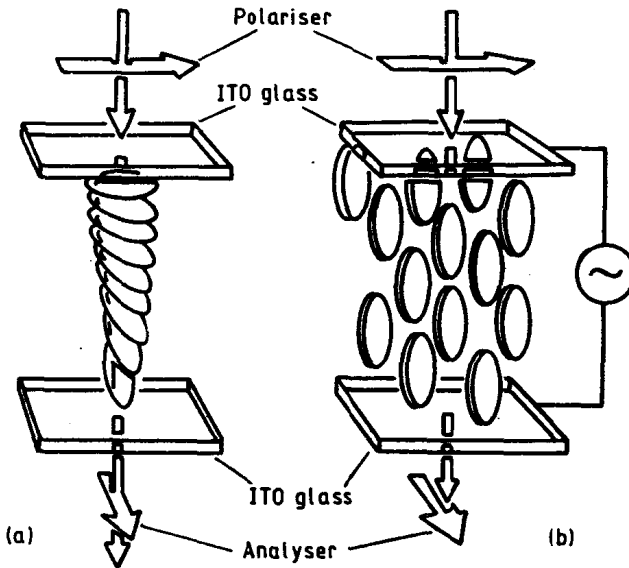
Figure 5. Freedericksz transition in a twisted nematic
LCD; (a) left-hand side $V < V_c$, (b) right-hand side $V > V_c$.

be transmitted if the device is placed between crossed polars. If a
voltage greater than threshold is applied, the twisted structure is
distorted as shown in figure 5(b); the guiding of plane polarised light
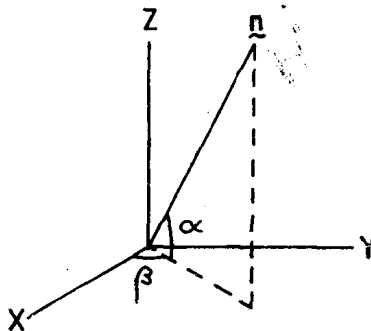is then lost and, with crossed polars, light is no longer transmitted.

Figure 6. Definition of director tilt and twist angles
α and β respectively. The plane of the cell is parallel
to z = 0.

Taking the tilt angle α and the twist angle β as coordinates (figure 6), equation (3.6) becomes

$$d^{-1}\int_0^d \delta(u_d + u_E)dz = \tfrac{1}{4}(\pi/d)^2[K_{11} - \tfrac{1}{4}(2K_{22} - K_{33})]\alpha_m^2$$

$$- \tfrac{1}{4}\varepsilon_0\Delta\varepsilon(D_z/\varepsilon_0\varepsilon_\perp)^2\alpha_m^2 + O(\alpha_m^4) \qquad (3.9)$$

where the lowest order term due to distortion from uniform twist is ignored since it is non-negative. Thus, exactly as in the parallel aligned cell, the Freedericksz transition is driven by distortion of the tilt profile, with a critical voltage

$$V_c = \pi[\{K_{11} - \tfrac{1}{4}(2K_{22} - K_{33})\}/\varepsilon_0\Delta\varepsilon]^{\tfrac{1}{2}} . \qquad (3.10)$$

The threshold given in equation (3.10) is the voltage at which the director begins to tilt, and the order parameter α_m becomes non-zero. However, the guiding of plane polarised light is not lost until the director tilt has become significant, causing the <u>optical</u> threshold to be
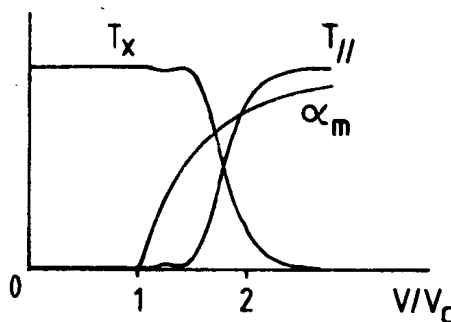

Figure 7. Schematic plots for a twisted nematic LCD of variation with RMS voltage V of tilt angle at centre of cell, α_m, and transmission with parallel polars $T_{/\!/}$ and with crossed polars T_X.

at a higher voltage. Figure 7 shows schematically the variations with RMS voltage of α_m and transmission with both parallel and crossed polars. The curves shown in figure 7 are for rays propagating normal to the cell. The optics of the twisted nematic are complex; it is evident from the symmetry group that they are not even cylindrically symmetrical. Rays at an angle