

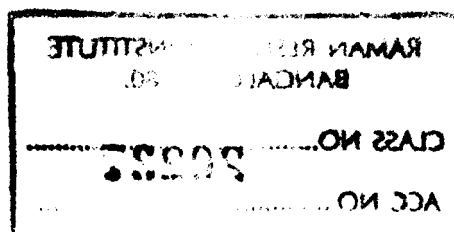
On the Elastic and Dynamical Properties of Biaxial Liquid Crystals

by

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Thesis submitted to the Jawaharlal Nehru University
for the degree of Doctor of Philosophy

1998



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DECLARATION

I hereby declare that this thesis is composed independently by me at the Raman Research Institute, Bangalore, under the supervision of Prof. G. S. Ranganath. The subject matter presented in this thesis has not previously formed the basis of the award of any degree, diploma, associateship, fellowship or any other similar title.



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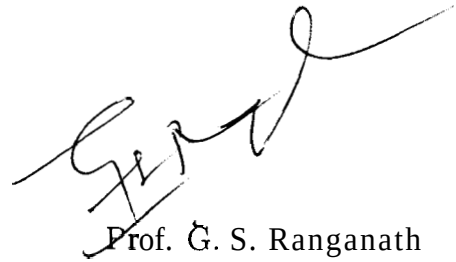
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CERTIFICATE

This is to certify that the thesis entitled On the Elastic **and** Dynamical Properties of Biaxial **Liquid** Crystals submitted by Sreejith Sukumaran, for the award of the degree of DOCTOR OF PHILOSOPHY of Jawaharlal Nehru University is his original work. This has not been published or submitted to any other university for any other degree or diploma.



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I remember my teacher, the late Mr. Thomas Mathai, who introduced me to the subject of Physics.

I dedicate this thesis to my family - they might not know what I did for this thesis, but they know me and that certainly means a lot to me.

CONTENTS

Preface	i
1. INTRODUCTION	1
1.1 Topological defects in nematics and smectics	5
1.1.1 Line disclinations	7
1.1.2 Point disclinations	12
1.1.3 Kinks or walls	17
1.2 Hydrodynamics of liquid crystals	21
1.2.1 Nematodynamics	21
1.2.2 Dynamics of smectics	22
2. ELASTIC INSTABILITIES IN BIAxIAL NEMATICS	26
2.1 Introduction	26
2.2 Elasticity of biaxial nematics	27
2.3 Director distortions due to rigid anchoring	29
2.4 Instabilities in the presence of a magnetic field	33
2.5 Formulation of a theory of elasticity of biaxial nematics	37
3. TOPOLOGICAL DEFECTS IN BIAxIAL NEMATICS	44
3.1 Introduction	44
3.2 Nonsingular defect structures	46
3.3 Singular hybrid disclinations	51
3.4 Defects in droplets	59
4. HYDRODYNAMICS OF SMECTIC-C	64
4.1 Introduction	64
4.2 Hydrodynamic variables	66
4.3 Conservation laws	67
4.4 Constitutive equations	70
4.4.1 Free energy and dissipative part of the stress tensor	73
4.4.2 Linearized equations	76

5. DYNAMICS OF THE UNIFORM STATE OF SMECTIC- C	80
5.1 Introduction	80
5.2 Inlet section: effects of permeation	80
5.3 Smectic Hall effect	84
5.4 Uniform state in a rotating field	86
6. HYDRODYNAMIC INSTABILITIES IN SMECTIC- C	96
6.1 Introduction	96
6.2 Field induced instabilities	97
6.2.1 Effect of C_2	99
6.2.2 Dynamic response	101
6.3 Flow induced instabilities	105
6.3.1 Shear plane normal to the layers	106
6.3.2 Shear plane parallel to the layers	108
7. DYNAMICS OF KINKS IN SMECTIC- C	113
7.1 Introduction	113
7.2 Theory	114
7.3 Instability of a 2π kink	118
7.4 Velocity selection	120
7.4.1 Tilted non-rotating oscillating field	123
7.4.2 Non-oscillating field rotating in a plane normal to the layers	129
7.5 Kinks in a field rotating about the layer normal	133
7.5.1 Dynamics of kinks in a non-oscillating field rotating about the layer normal	134
7.5.2 Dynamics of kinks in an oscillating field rotating about the layer normal	136
7.5.3 Metastable kinks in a field rotating about the layer normal	140
7.5.4 Lattice of kinks	141
7.6 Remarks	141

Preface

Liquid crystals are states of condensed matter between a crystal and an isotropic liquid. They are characterized by a preferential orientation of their highly anisotropically shaped molecules. They may also possess positional order. Further, they can be of uniaxial or biaxial symmetry. A biaxial liquid crystal possess two directions of optical isotropy and its electric or magnetic susceptibility tensor is a triaxial ellipsoid. This thesis deals with the elastic and dynamical properties of biaxial liquid crystals, in particular biaxial nematics and smectic-*C*. Biaxial nematics do not possess positional ordering. The most symmetric biaxial nematic has orthorhombic symmetry with the preferential alignment of the molecules described by a triad of unit vectors also called directors. About each of these, the medium has a 180° or a two-fold rotational symmetry. In the case of smectic-*C*, the constituent molecules are stacked in layers and preferentially aligned at an angle with respect to the layer normal. It has monoclinic symmetry with a two-fold axis perpendicular to this direction and a mirror plane perpendicular to the two-fold axis. In chapter one, the elastic and dynamical properties of liquid crystals are briefly reviewed. This provides the background material for the rest of the thesis.

In a uniform biaxial nematic (also called a single domain), the triad of directors has the same orientation everywhere. However, in the presence of external electric or magnetic fields, or different director anchorings at the surfaces, the orientation of the triad varies from point to point. These distortions are curvature distortions in the three director fields and can be looked upon as elastic director distortions. These elastic distortions in the bulk of a biaxial nematic with orthorhombic symmetry can be described effectively within the framework of a continuum theory, and there are 12 curvature elastic constants. By contrast, normal nematics are uniaxial

and are described by one director field and three curvature elastic constants. As a result, in biaxial nematics, the equations of equilibrium are complex. This is one of the reasons why this exotic phase has remained largely unexplored. In this thesis, attention has been drawn to elastic instabilities and topological defects in biaxial nematics, and they are studied in chapters two and three respectively. Elastic instabilities can arise when the existing orientation of the orthonormal triad of directors spontaneously undergo changes. This can happen due to external electric or magnetic fields. Also, the inherent biaxiality of the material will generally lead to a coupling between distortions in the three different director fields and this can lead to elastic instabilities. The general problem with the consideration of the 12 elastic constants is mathematically cumbersome and intractable. On the basis of a new formulation of the continuum elastic theory, we justify an approximation that gives an intuitive understanding of the elasticity of biaxial nematics and also, makes the mathematics tractable. Some of the significant results presented in chapter two are:

- A theory of elasticity of biaxial nematics which incorporates a variable degree of orientational order parameter has been formulated. On the basis of this theory, we can justify the use of a simplified model of elasticity in which the distortions are described by only three elastic constants and still, this approximation preserves effects due to inherent biaxiality. This theory naturally allows for a description of topological defects with accompanying order parameter variations.
- In a confined geometry, with a uniform twist deformation of two of the three directors, we find an elastic instability with one of the directors lifting out of the plane of the plates. This instability can happen at a threshold of total twist less than 90° and is therefore easily demonstrable in the laboratory. On the contrary, in a similar instability in a uniform twisted uniaxial nematic, the

threshold of total twist is close to 180° which is unattainable experimentally.

- In view of its symmetry, it is possible to have a geometry which imposes a twist in one of the directors with the anchoring of the other two directors being different on two parallel plates. In such a situation, a uniform twist does not exist. The inherent biaxiality is responsible for this nonuniformity in the twist and the nonuniformity increases with increasing difference between the three elastic constants. There is no counterpart of this situation in uniaxial nematics.
- Elastic instabilities that arise due to external electric or magnetic fields have been considered on the basis of a Landau theory and phase diagrams of structural changes have been obtained which indicate that there can be first order and second order phase boundaries. It should be remembered that in the case of uniaxial nematics, there can only be second order phase boundaries. The richer phase diagram allows for more complex elastic instabilities. For example, in one particular geometry which leads to only two structures in the case of uniaxial nematics, there will be four distinct structures in biaxial nematics.

Just as in uniaxial nematics, topological defects exist in biaxial nematics. There are four stable line disclinations in such media, viz. three distinct "half-strength" disclinations with winding number or strength, $s = \frac{1}{2}$, and a disclination line of strength $s = 1$. These can be wedge, twist or hybrid disclinations; the last having both wedge and twist components. It can be shown that a disclination line of odd-integral strength is singular, and would split itself into half-strength disclination lines. Hence, it is natural to expect only half-strength disclinations in biaxial nematics. But, nonsingular disclinations are also permitted in biaxial nematics. In chapter three, the structure, energetics and interaction of disclinations have been explored in detail. A few of the interesting conclusions are:

- It is possible to have a nonsingular hybrid disclination which would be energetically preferable to a half-strength disclination. Thus, a biaxial nematic need not only have half-strength disclinations.
- The interaction between hybrid disclinations is determined not only by sign and strength of its twist and wedge components but also by the biaxial elastic anisotropy. In the case of the interaction between a pure wedge disclination and a hybrid disclination with wedge components of the same sign, there could be attraction or repulsion depending upon the strength of the twist component and the elastic anisotropy. Similarly, in an interaction between hybrids when only one of the components are of opposite sign, there can be either attraction or repulsion.
- When one of the triad of directors is normal to the surface of a spherical drop of biaxial nematic, we find that there are two possible defect structures for the drop. In the first configuration, there is a tetrahedral arrangement of four half-strength disclination lines emanating from the centre of the drop. In the second configuration, there are no singularities in the director fields anywhere except at a point on the surface of the drop. Such a configuration is often called a boojum. Our calculations imply that for smaller drops, the tetrahedral disclination structure is the lower energy state. But, for larger drops, the boojum would be of lower energy for the same boundary conditions.

In the remaining four chapters of the thesis, attention has been paid to the other biaxial liquid crystal. *viz.* smectic- C . Firstly, the hydrodynamics of smectic- C is presented. The first formulation of the hydrodynamics of smectic- C as developed by Martin, Parodi and Pershan (MPP) has the essential features of both the permeation process and elastic deformations in layer spacing but does not allow for large

scale distortions. It can be used to study only small fluctuations and wave propagation. More recently, Leslie, Stewart and Nakagawa formulated a nonlinear theory which allowed for large scale distortions of the director. This theory has been extensively used in recent times to describe the orientational director dynamics and flow behaviour. But, in this theory, the permeation process and layer elasticity have not been incorporated. Another drawback of this theory is that it does not reduce to the linearized theory of MPP. In chapter four, a complete nonlinear continuum theory of the isothermal hydrodynamics of smectic-C, with the advantages of both these theories, is formulated. Using an asymmetric stress tensor and Onsager's reciprocity relations, it is found that the entropy production embodying dissipative torques and shear stresses is described by 16 viscosity coefficients, and a permeation constant arising due to the dissipative motion normal to the layers of smectic-C. The linearized version of this theory does reduce to that of MPP. This theory incorporates coupling between the different hydrodynamic variables, viz. velocity, layer spacing and orientation of the director. Here, two of the salient features of this theory are pointed out.

- It is a covariant description in the sense that the constraints of the system have been incorporated and the theory is applicable for large scale distortions.
- The dynamics of the director is described by an equation similar to the Oseen equation in uniaxial nematics. The formulation of the constitutive relations using an asymmetric stress tensor not only leads to a more intuitive understanding of the dynamics of the director, but also highlights the nature of the permeation process in a continuum description. We consider situations not involving creation of dislocations, *i.e.*, the number of layers is conserved, and surface torques and layer depinning are assumed to be absent. Then, the permeation process wherein there is a relative motion of the fluid particles with

respect to the layers always leads to a distortion of the layers.

In the next three chapters, attention is drawn to the implications of this theory in three situations, *viz.* (i) flow and director dynamics of the uniform state, (ii) field and flow induced structural and hydrodynamic instabilities; and finally (iii) dynamics of topological walls or kinks. The dynamics of the uniform state is discussed in chapter five. Poiseuille flow and director motion in a rotating field are considered. Some of the important results are:

- In the Poiseuille flow of ordinary fluids between parallel plates, the fluid flow attains steady state over a length called the inlet section. This length is comparable to the separation between the plates and is proportional to the main flow velocity. In the case of smectics, in the same geometry but with layers slipping past one another and parallel to the plates, permeation flow exists in the inlet section. As a result of this, it is found that the length of the inlet section becomes independent of the main flow velocity. Further, the length of the inlet section turns out to be many times the separation between the plates. For example, when the separation between the plates is 0.001 cm, this length is nearly 10 cms. Therefore, actual experimental results in short tubes are not really representative of the steady Poiseuille flow.
- In the case of Poiseuille flow with the layers in the shear plane, *i.e.*, the layers are fixed and fluid flows in the layers, a transverse pressure gradient develops in order to maintain flow in the layers. Nematics with molecules at an angle to the shear plane also exhibit a similar effect. This is often referred to as "nematic Hall effect". Unlike the situation in nematics, the smectic Hall *effect* exists even when there is no external electric or magnetic field to tilt the molecules out of the shear plane and in this sense it is peculiar to smectic-C.

- When a constant magnetic field is rotated about the layer normal, the behaviour of the system falls in one or the other of two regimes. In the synchronous regime, the director follows the field at an angle constant in time. This happens as long as the rotational frequency is below a critical value. When the frequency of rotation is beyond the critical value, the system goes over to an asynchronous regime in which the director does not follow the field with a constant angle but exhibits a stick-slip process wherein the director follows the field for a long interval of time at nearly a constant angle and then suddenly slips through 2π over a short interval of time. The phase-slip frequency is a continuous function of the frequency of rotation. When the rotating magnetic field is also oscillating, the frequency at which this phase-slip occurs is always a rational fraction of the frequency of the oscillating field. The dependence of the phase-slip frequency on the rotational frequency is rather like a 'Devil's staircase'.

In chapter six, a linear stability analysis of instabilities that occur under the destabilizing influence of an external field is described. In particular, we consider a geometry with both the layers and the external field perpendicular to the plates at which the director is rigidly anchored parallel to the plates. Thus, the orientation due to the field alone is in competition with the orientation imposed by director anchoring at the walls. Then, it is found that beyond a threshold value of the field, there is an instability to a deformed configuration. Instabilities in flow geometries with the flow along the layers have also been considered. The shear plane can be either parallel or perpendicular to the layers. In both field and flow induced instabilities, previous investigations had ignored permeation. Though similar instabilities are possible in uniaxial nematics, the analysis brings out new features in the instabilities in this biaxial liquid crystal in these geometries. Some of the new results are:

- In the case of field-induced instability, attention has been paid to the effect of an external magnetic field, applied perpendicular to the plates and along the layers, on a single domain of smectic-C with the layers normal to the plates and the director parallel to the plates. With the onset of instability, there arises a curvature distortion of the director and the smectic layers with fluid flow along and normal to the layers. Interestingly, at high fields, the director distortion near the plates and near the centre can even be of opposite senses. Further, the transient flow along and normal to the layers can become comparable in magnitude.
- When the shear plane is normal to the layers and the director lies in the shear plane, there exists a uniform steady state in which the projection of the director onto the layers is either parallel or antiparallel to the fluid velocity. If the director is anchored to the plates and the layers are parallel to the plates, an instability to a twisted director configuration arises above a threshold shear. With the onset of this instability, there is also a steady transverse flow. At this point, it should be noted that this is peculiar to smectic-C and there is no analogue of such a situation in uniaxial nematics. In uniaxial nematics, the uniform steady state is permissible only when the director is aligned at an angle, with respect to the flow, in the shear plane. This particular angle of alignment is due to flow alignment, and it is determined by the ratio of two viscosity coefficients associated with viscous torques.
- Another flow geometry considered is shear flow with the shear plane parallel to the layers. In this geometry, in the absence of anchoring at the walls, the director will everywhere be at the same angle with respect to the flow direction. If the director is aligned at the walls at this very angle, then we expect the flow to be stable. But a permeation induced instability occurs above

a threshold shear and, it involves a distortion in the director field accompanied by distortions of the layers due to transverse permeation flow.

Finally, chapter seven deals with the dynamics of kinks in smectic-C. Under a sudden destabilization, systems in a wide class of problems respond by forming a front which propagates from a stable to an unstable or a metastable state. And the problems of interest are in the formation of a unique propagating kink, and whether the system selects its speed from many possible speeds. The dynamics of twist kinks have been considered to address these issues. A π twist kink has a 180° twist of the director connecting two uniform states in an external field. In particular, the dynamical effects of oscillating and rotating magnetic fields have been studied. Some of the new and interesting results are:

- Recent investigations of others. in these systems, indicate that there could be chaotic instabilities in π twist kinks when an oscillating electric or magnetic field is applied at an angle to the layers. With a reanalysis of this problem. an entirely different result is obtained. It is found that the dynamics is not chaotic but periodic. In addition, it is shown that there is a crossover in velocity selection. This means that the structure and velocity of a π kink are not described by a single function of the tilt angle of the field. This is probably the first example of a system choosing between families of fronts which are not uniformly travelling, but varying periodically in time.
- Velocity selection is seen even when a non-oscillating field is rotated in a plane normal to the layers. Strangely! in this geometry the kink develops a drift velocity over and above an oscillatory motion and the drift is dependent on the sense of rotation of the field. A simple consequence of this is that a 2π kink could be in a bound state or it could split into two repelling π kinks depending on the sense of rotation.

- In the asynchronous regime of a field rotating about the layer normal, it is found that due to the effects of a natural finite relaxation time of the director and the rapid variation due to a "stick-slip" process, the width and velocity of the kink are aperiodic in time.
- The dynamics of kinks in an oscillating field rotating about the layer normal is even more complex. The kink motion is basically decided by a phase-slip frequency which varies discontinuously with the frequency of rotation of the field. Its dynamics has both periodic and aperiodic features being very sensitive to the parameters of the geometry.

The studies reported in this thesis have led to many more important problems which are being pursued. Many of these investigations were carried out in collaboration with the thesis supervisor. Part of these studies have been already published in the following papers.

1. *On some elastic instabilities in biaxial nematics*, Sreejith Sukumaran and G. S. Ranganath, Journal de Physique II, vol. 7, (1997) p. 583 - 601
2. *Dynamics of kinks in Smectic-C Liquid crystals in periodically varying external fields*, Sreejith Sukumaran and G. S. Ranganath, Physical Review E, vol. 56, (1997) p. 1791 - 1803
3. *Hydrodynamics of Smectic-C Liquid crystals : Field and flow induced instabilities in confined geometries*, Sreejith Sukumaran and G. S. Ranganath, Physical Review E, vol. 57, (1998) p. 5597 - 5608