Some Studies on Defect Lattices in Liquid Crystals

by

Andal Narayanan

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

1996

Raman Research Institute
Bangalore 560 080

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.

(Prof. G.S. Ranganath)
Raman Research Institute

Bangalore.

(Andal Narayanan)

CERTIFICATE

This is to certify that the thesis entitled **Some Studies on Defect Lattices in Liquid Crystals** submitted by Andal Narayanan, for the award of the degree of DOCTOR OF PHILOSOPHY of Jawaharlal Nehru University is her original work. This has not been published or submitted to any other university for any other Degree or Diploma.

N. Kumar

Prof. N. Kumar

(Centre chairperson)

Prof. G.S. Ranganath

(Thesis Supervisor)

Director
Raman Research Institute
Bangalore.

ACKNOWLEDGEMENTS

It is a great pleasure to thank my thesis supervisor Prof. G.S. Ranganath. Though the years passed by in a whirlwind of activity, looking back now, I can fully appreciate the forethought and wisdom which has gone in, while he took decisions. As a teacher, he has helped me learn to look at things, to a first order, from the viewpoint of a curious outsider. I indeed have benefitted immensly under his guidance.

I thank Prof. K.A. Suresh for the immense patience with which he critically went through all our papers not to mention, the thesis as well. His constructive comments had always had the effect of improving the presentation.

A right ambience is an essential criterion in a student's life. I thank Prof. J. Samuel for the stimulating discussions I had with him. From the standpoint of a 'curious outsider', I have benefitted from the discussions Prof. R. Nityananda had with others - 'They also learn who stand and listen'. I acknowledge with warm regards the e-mail discussions with Prof. R.B. Meyer and Prof. Lui Lam. In particular, the discussions with Prof. R.B. Meyer have been illuminating.

Dr. P.B. Sunilkumar was very helpful during my initial computing stages of the 'Berrimann' problem. He and Dr. Yuvraj Sah were first-aid centers, in those days when everything seemed to go wrong everytime. I thank them for their patience.

I also thank Prof. N. Kumar, Director, Raman Research Institute for his interest in my work.

The facilities provided by the library, not only in terms of a very good collection of books but also with a sunny staff who lent a patient ear to many queries, should be one of its kind in India. Many a murky mood has been broken after a few hours in the library. I thank Mrs. Girija and her colleagues for wonderful time spent in the library. Special thanks to Mr. Hanumappa and Mr. Chowdappa for photocopying the thesis. I acknowledge with thanks the excellent computing facility at the institute and the ready help rendered by the computer staff. In particular, I would like to thank Mr. Rajsekhar, Mr. Ramadurai and Ms. Ashima.

Most of the precious time could well have been wasted in bureaucratic hassles. I thank the administrative staff of RRI. with special thanks to Mr. Krishna who made everything look very simple.

My seniors, batch mates and juniors have made my stay at RRI a very enjoyable experience. I have enjoyed the versatality and wit of Ms. Usha during my formative years here. I thank Dr. Ramesh, Ms. Lakshmi and Ms. Seethalakshmi for coloring all the dull weekends.

Serious discussions on completely unknown topics could not have been made possible but for an immediate response from Dr. T.K. Sridharan. He and Ramadurai have always been eager to pursue a deductive approach even on unheard topics.

Technical and non-technical discussions with Ms. Sushan Konar can always be looked back with relish.

Mr. Sreejith's skepticism and uncompromising attitude in Science has made an impact. Mr. Vikram and Ms. Sobha were both instructive-one for the extreme patience and the other for a down to earth approach. I take this oppurtunity to thank all my hostel-mates with whom I interacted during my stay there.

I have been very fortunate to have friends who have always been with me when I passed through various phases of my life. Ms. C. Indrani and I had had discussions, arguments and altercations on almost everything known to both of us. I can only look back our years together as one long trip of learning and growing.

Ms. Nimisha Kantharia was invaluable in her companionship. Her deliberate good humour, pleasant nature and sympathetic understanding of situations has gone a long way in making this thesis a reality.

As a stimulating friend and a mentor, Ms. Uma Gorti has had a very subtle and deep influence on my way of thinking about life. Her sharp intellect and unorthodox thinking has made a world of difference.

Ms. Sarala has made the last few months of stressfull period less painful. Her stoicism and perseverance always lifted my spirits.

My deep gratitude to my parents who instilled in me the right perspective. But for their constant encouragement neither the ups nor the downs could have been scaled. To my grandmother I owe a lot for her just being there always.

Last but not the least I would like to thank Mr. T.P. Saravanan for everything.

Contents

PREFACE

Liquid crystals exhibit beautiful optical textures. These are an aglomeration of topological defects. A study of such defects has become an active area of investigation in recent times, in view of their interesting structures and properties. Further their study has lead to a better understanding of phase transitions in such condensed matter systems.

A topological defect in an ordered phase is intimately related to its symmetry elements. For example, an edge dislocation in a crystal is a permitted topological defect that is consistent with its translational invariance. One can also consider defects called disclinations, in crystals that are consistent with their rotational invariance. However, these defects would have prohibitively large energies. On the other hand in liquid crystals, disclinations are associated with very small energies and they exist on their own in these systems. In addition, since a liquid crystal is both dielectrically and diamagnetically anisotropic, new defect states are permitted in the presence of an external electric or magnetic field. Helfrich predicted one such defect state. This is similar to the Bloch and Neel walls of the magnetic systems. For instance, in a *Nematic* liquid crystal, the director which denotes the average orientation of the molecules can either be parallel or anti-parallel in an external electric or magnetic field. A *Helfrich Wall* in a nematic liquid crystal is a non-singular defect state connecting these two director orientations. These states can be connected through a splay, bend or twist distortion in the director field. then the wall is referred to as a splay wall, a bend wall or a twist wall respectively. The director undergoes a rotation of $180°$ or π as we cross the Wall from one uniform state to another. This distortion is confined to a narrow region in space which corresponds to the width of the Wall. Incidentally, these structures have come to be designated in liquid crystal literature as *Planar Solitons*. This thesis reports some theoretical investigations on the structure, dynamics and optics of solitons and soliton lattices in liquid crystals. The introductory chapter reviews the structure and energetics of single soliton states. During these studies some new heither to unrecognised, static soliton states were found to be permitted defects in liquid crystals. Discussions on some of these are also included in this chapter.

A Cholesteric liquid crystal is characterised by a spontaneous rotation of the director perpendicular to itself. This gives a lattice like periodicity to the structure with a period called the *pitch* within which the director rotates through 360° or 2π. It should be remembered that there is no density modulation occuring in the system. A nematic state can be obtained from such a cholesteric liquid crystal by the application of a magnetic field. We have undertaken a study of the structure and energetics of planar solitons in this nematic state. They are found to be very different from their counterparts in ordinary nematics. This forms the subject matter of the second chapter. Some new results obtained are:

> It is well known that in a field perpendicular to the twist axis of a cholesteric the structure goes over to an unwound state (nematic like) only above a threshold field strength. In this nematic twist, anti-twist and bend solitons are not only permitted states but they also connect the same uniform director orientations. An anti-twist soliton which has a sense of twist opposite to that of the cholesteric can become energetically favourable compared to the bend soliton. Further in certain field regimes the bend soliton and an anti-twist soliton can have comparable energies leading to the existence of a new type of disclination line connecting these two solitons. Interestingly, in a normal

nematic both these solutions are generally not permitted. This is because the bend elastic constant for the director is always higher than the twist elastic constant.

Generally, a cholesteric can also go over to a nematic state in a field parallel to the twist axis. In such nematics a new giant soliton is found to be a permitted defect state. The width of this twist soliton increases on decreasing the field and its energy continuosly decreases and goes to zero at the critical field H_C. The entire structure is then transformed into a cholesteric.

Unwinding of a cholesteric in a magnetic field is the most thoroughly studied of chiral-achiral transitions in liquid crystals. Such field induced transitions between other chiral-achiral liquid crystals is an important area of research. This has so far only been studied in a limited number of situations. We consider this afresh in some generality. Cholesteric and chiral smectic liquid crystals (a liquid crystal with an one dimensional density modulation) can be doped with magnetic grains to make ferrocholesteric and chiral ferrosmectic liquid crystals respectively. In these systems, the magnetisation spirals about the twist axis. It may be pointed out that, the term chiral or achiral refers to the presence or absence of a macroscopic twist in a particular liquid crystal irrespective of the chirality occuring at the molecular level. These studies reported in the third chapter, has lead to many interesting results. Some of them are briefly presented here.

A ferrocholesteric in a magnetic field parallel to the twist axis, goes over, on increasing the field, to an achiral state through a gradual tilt of the director and becomes nematic like at a critical field. On decreasing the field the transition reverts back to the chiral state. Interestingly, this reverse transition takes place through a generation of non-singular disclinations. This transition can even a have reentrant behaviour. Very similar transitions take place in chiral ferrosmectics also. But here, the achiral to chiral transition is mediated by singular disclinations. Further, the transition point is dependent on the direction of the magnetic field.

The study of such transitions in these systems in crossed electric and magnetic fields have also lead to important results.

A cholesteric in a crossed electric and magnetic field perpendicular to the twist axis goes over to an achiral state through the formation of a twist soliton lattice i.e., a periodic array of single π twist solitons. However, this lattice on increase of electric or magnetic field goes over to an achiral state aligned either along the magnetic field or perpendicular to it. On the other hand a ferrocholesteric, in the same geometry goes over to the achiral state through formation of a 2π twist soliton lattice. Further each such soliton has a finer structure in the nature of a split soliton i.e., it is made up of two solitons. This split is symmetrical in systems with positive diamagnetic anisotropy and asymmetric in systems with negative diamagnetic anisotropy.

Field induced chiral-achiral transitions in ferrosmectics are equally interesting in crossed fields but with the magnetic field along the twist axis and electric field perpendicular to it. Again the achiral phase is reached through the formation of a twist soliton lattice. One finds in this case a rich phase diagram with many important features like the phenomena of reentrance, tricritical and Lifshitz points.

The single solitons and soliton lattices considered till now, are all static structures. A soliton can be made to move from one stable state to another if it connects director orientations of different magnetic potential energy. It also moves when the symmetry in potential energy is broken within the core of the soliton. This can be effected by rotating the field in which we have the soliton. We consider in the fourth chapter situations in which both factors viz., the core asymmetry and the base state asymmetry are operational. The velocity of the soliton is dependent, obviously, on the driving force which is propotional to the frequency of rotation of the magnetic field. It is well established that this velocity diverges at a certain value of the driving force. However a soliton which propagates from a stable state to an unstable state has very different dynamical properties. Also such a soliton is associated with an exponentially decaying periodic director distortions. We call them as solitons with a tapered lattice. There are two kinds of tapered lattices depending upon the director distortions. In a twist tapered lattice only a twist distortion exists. On the other hand, a splay-bend distortion alone exists in a $splay-bend$ tapered lattice. In all such solitons one side is in a uniform state while the other side has a oscillatory structure called the tapered lattice. Incidentally, a twist tapered lattice can also be looked at as an alternate stack of right handedly and left handedly twisted structures. The magnitude of the twist decreases as we go down the lattice. In a similar fashion, the splay-bend tapered lattice can be looked upon as an alternate stack of splay-rich and bend-rich regions. The properties of these two kinds of tapered lattices are very different from those of solitons which connect stable states. Another periodic structure which exhibits a very different dynamics from that of a usual single soliton is a multi-soliton lattice. This is made up of an array of single solitons.

The dynamics of tapered lattices and multisoliton lattices in nematics and ferronematics form the subject matter of the fourth chapter. A few of the important results obtained are given here.

> It is well known that a soliton with a tapered lattice does not have a unique velocity. There is a range of velocities for every value of the driving force. Even in this range of velocities the tapered lattice structure of the soliton will or will not exist depending upon the velocity. We find the velocity of propagation of these solitons to be very different from that of single solitons. In particular there is no velocity divergence. Further both the rotational frequency and the strength of the field affect considerably the structure of the tapered lattice.
>
> In the case of multi-solitons have some unusual features are found. The velocity is a single valued function of the rotational frequency. In this respect it resembles single solitons connecting stable states. But unlike the case of single solitons, the velocity does not diverge. It increases monotonically in the beginning, reaches a maximum and then falls rapidly to zero at a critical value of the driving force (rotational frequency). Interestingly, the period of the multisoliton lattice diverges at very small values of the rotational frequency. This appears to explain the experimental observation of Migler and Meyer who find the spacing of a spiral lattice to diverge as the driving force is decreased.

The structure of both solitons and soliton lattices can be studied using optics as a tool. Due to their birefringence, liquid crystalline materials show rich optical features. Optical properties have been worked out both in the reflection and in the diffraction modes. In this context it might be stated that a Twist Grain Boundary Smectics is rather structurally similar to a twist soliton lattice. In view of this fact, its optical properties have also been studied. In chapter five, studies on these defect lattices in the reflection mode have been presented. In the next chapter is considered their diffraction properties. In the last chapter, the unusual effects that can exist in defect lattices due to the presence of optical absorption are highlighted. Some of the important results arrived at these optical studies are:

In the reflection mode there are multiple reflections for twist grain boundary smectics, even at normal incidence. We not only get a strong reflection at a wavelength corresponding to the optical period of the structure but also at its harmonics. Some of these reflections are either right circularly polarised or left circularly polarised while other reflections are polarisation insensitive. However, in the band corresponding to the optical period, the handedness of the strongly reflected circular state is seen to have same sense as that of the structure.

In samples with finite thickness, there is a phase difference between the transmitted right and left circularly polarised light, in bands where one of these polarisations get strongly reflected. This leads to optical rotation. As in cholesterics, this optical rotation is anomalous inside the band. That is, it not only goes to very high values inside the band but also changes sign within it.

Reflections at normal incidence from a twist tapered lattice also leads to many reflection bands corresponding to harmonics of the fundamental period. Since this structure can be looked upon as an alternate stack of right and left handed structures, it is natural to expect every band to reflect both right and left circular light. But these polarisations are reflected with unequal intensities in bands which are even harmonics of the fundamental. Also in these bands we get anomalous optical rotation. Another unusual property of the tapered lattice is that the reflection coeffiecient of light traveling along the twist axis in one direction is different from that traveling in the opposite direction.

On the other hand, a splay-bend tapered lattice has strong reflection only at even harmonics of the fundamental. Further these bands reflect only linearly polarised light which is polarised along the direction of uniform director orientation. This structure also exhibits asymmetric reflection of light for propagation along and opposite to the direction of director modulation.

Diffraction mode has also many unusual features. The diffraction pattern has been worked out for twist grain boundary smectics and tapered lattices for light incident normal to the direction of lattice modulation and the incident light is assumed to be linearly polarised. Some interesting results in this case are:

In twist grain boundary smectics, the diffraction pattern is symmetric with respect to the zeroth order. There is no diffraction for light polarised parallel to the twist axis in the case of some twist grain boundary smectics.

On the other hand diffraction pattern is asymmetric both in twist tapered and splay-bend tapered lattices. Further, there are specific linear polarisation states of incident light for which diffraction will not take place.

When these defect lattices are doped with dyes or if they have intrinsic broad absorption bands in the visible we find many other novel optical features both in the reflection and the diffraction mode.

For example, uniformly absorbing twist grain boundary smectics exhibits Bormann effect. As a result, in some reflection bands which reflect a particular (right/left) circularly polarised light, the transmittance for the reflected state is greater than the transmittance of the other (left/right) circular light which experiences an average absorption. In some bands, this anomalous transmission is present at both the edges of the reflection band and in some others it is totally absent.

Absorbing twist tapered lattice exhibit asymmetric reflection bands while the splay-bend reflection lattice has symmetric reflection bands. In addition the reflection asymmetry already referred to in the non-absorbing case, is all the more pronounced here.

In the diffraction mode too we find some interesting results. For instance we find asymmetric diffraction pattern in some twist grain boundary smectics while it is symmetric in other twist grain boundary smectics. Also, generally, there is no diffraction for polarisation parallel to the twist axis in uniformly absorbing twist grain boundary smectics. On the other hand, in a non-uniformly absorbing system where only the grain boundaries are absorbing, we find diffraction for this polarisation also. A study of this pattern leads to a direct estimate of the thickness of the smectic blocks. Further, twist grain boundary smectics which exhibited asymmetric diffraction when they were uniformly absorbing will have a symmetric diffractiort pattern when only the grain boundaried absorb.

These studies on defect lattices have further led to many other investigations which are being pursued.

Most of the problems considered in this thesis were suggested by the thesis supervisor Prof. G.S. Ranganath and were worked out by the author. Some of the important results of these studies have already been published in the following papers.

(1) **On the Optics of Twist Grain Boundary Smectics.,** N. Andal and G.S. Ranganath., J. Phys. 11France., ***5*** 1193 (1995)

(2) **Field Induced Chiral-Achiral transitions in Liquid Crystal.,** N. Andal and G.S. Ranganath J. Phys. 11France., 6 639 (1996)

(3) **Structure of planar solitons in nematic and smectic liquid crystals.,** N. Andal and G.S. Ranganath., Liquid Crystals., ***20*** 321 (1996)

(4) **Optics of some absorbing defect lattices.,** G.S. Ranganath, N. Andal and K.A. Suresh., International Conference *on* liquid crystals, *Kent state,* June *1996*.

(5) On Solitary waves in some liquid crystals N. Andal and **G.S.** Ranganath (in preparation)