

Chapter 1

Introduction

1.1 Liquid Crystals

Liquid crystal is a state of matter intermediate between positionally and orientationally disordered liquid and positionally and orientationally ordered crystal. The constituent molecules which form these phases are highly anisotropic in their shapes. When crystals made of such anisotropic molecules melt to form a liquid phase they generally go through an intermediate thermodynamic phase which is the liquid crystalline phase. This liquid crystalline phase is characterised by the presence of an orientational order and by the absence of crystalline order in one, two or three dimensions. In some compounds this loss in positional order can be brought about by a change in temperature. This results in thermotropic liquid crystals. In a multicomponent system, which comprises typically of rigid rod like polymers in a liquid substrate, liquid crystalline phases appear in a range of concentration of the component molecules. Such liquid crystals are called lyotropic liquid crystals.

Liquid crystals exhibit characteristic optical textures under a polarising

microscope. These textures are due to a collection of topological defects called *disclinations*, which are characteristic of liquid crystals. Most of the liquid crystalline phases are often identified by a study of such textures. Some of these defects exist naturally in a given sample of a liquid crystal. Disclinations can also exist in some liquid crystals in an external magnetic or electric field. Another natural topological object which can exist in a liquid crystal **but in the presence of an external electric or magnetic field is a defect state** called soliton. In some field induced phase transitions between different liquid crystalline phases, solitons play an important role. Apart from that they have unique elastic and hydrodynamic properties.

Defects in liquid crystals have close analogies with those found in other states of condensed matter like crystals, superfluids and magnetic systems. Thus a study of liquid crystal defects which are easy to undertake give an insight into defects in other systems which call for more sophisticated techniques.

In this thesis, we have undertaken a theoretical study on the elastic, hydrodynamic and optical properties of solitons. In particular, emphasis is on i) single soliton states with an associated lattice structure ii) solitons in a liquid crystal with a latent lattice structure and finally iii) a lattice of single solitons. All these we study in different contexts. In this introductory chapter, a brief review on a planar soliton is presented. This is necessary for the development of the subject matter of the thesis. This chapter also has some new results pertaining to single solitons, which were obtained in the course of our study. The second, third and fourth chapters concern themselves with the mechanical properties of these defects. This includes a study of the structure and propaga-

tion of single solitons and multisolitons. A simple way to probe the structures of many of liquid crystal defects is through optics. This is particularly important in our case as most of the defects of interest have periodic structures associated with them. Optical reflection and diffraction in such structures is the subject matter of the last three chapters.

1.1.1 Classification of liquid crystals

Liquid crystal systems are made of highly anisotropic rod like or disc like molecules which in general have no positional order. Hence the degree of freedom distinct from the localization of centre of gravity of an individual molecule constituting a liquid crystal, is its orientational order. A natural way to classify these systems is to start with a liquid with no positional order but with orientational order and impose positional order in one or two dimensions [1].

Achiral liquid crystals

Nematics

The simplest of liquid crystals are Nematics. In this liquid crystal there is no positional order in all the three directions but only a long range orientational order. Nematics can be made of both rod-like (or disc-like) molecules. They are all oriented on an average, along (or perpendicular to) a specified direction. This direction is denoted by an unit vector n called the "director" (Figure 1.1). The director is apolar i.e., $n = -n$ due to the existence of a plane of mirror symmetry perpendicular to it. This phase has uniaxial symmetry and belongs to the point group element $D_{\infty h}$. A nematic phase with a biaxial symmetry has also been realised

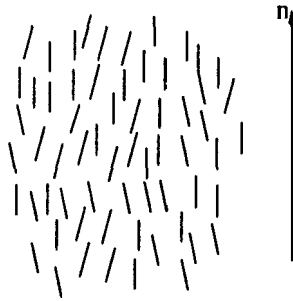


Figure 1.1: *Schematic representation of nematics*

in recent times.

Smectics

When a nematic like orientational order has also in addition a translational order along a particular direction, we end up with stacks of two dimensional liquid layers. That is, density modulation occurs in only one direction with a quasi long range lattice order. These systems are called smectics. The simplest of these liquid crystals is Smectic A (S_A) (Figure 1.2) with the point group symmetry of a nematic. Here the director n and the direction of the density modulation are parallel to one another. In other words the normal to the liquid layers and the director are in the same direction. In a smectic C (S_C) liquid crystal, the average orientation of the molecules in the liquid layer is at an angle to the layer normal. This liquid crystal is of monoclinic symmetry and belongs to the point group C_{2h} . Since the director n makes an angle with the layer normal, the projection of the director on to the layers is a well defined

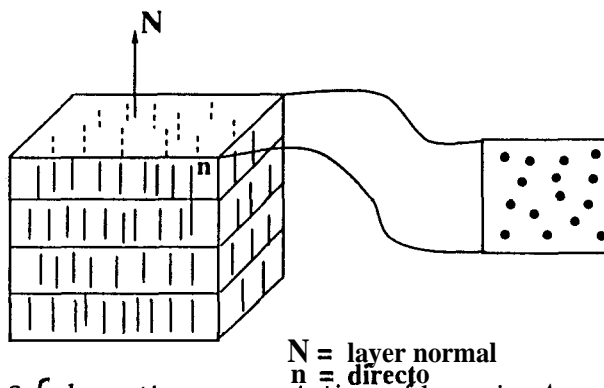


Figure 1.2: Schematic representation of nematic A

vector called the c . Obviously c is a polar vector. There could be even bond orientational order in the smectic layers. Such liquid crystals are called hexatic phases.

- Discotics

These are phases which have two dimensional positional order with liquid order in the third dimension. In general, this phase is made up of disc like molecules stacked with liquid like order. These stacks are arranged in a regular lattice. These are also called columnar discotics.

Some additional classification of liquid crystalline phases has also been made depending on the nature of the molecules forming the liquid crystalline phases. If the constituent molecules are non-centro symmetric (chiral), then we get some additional phases which are related to one or other of the above three categories.

Chiral liquid crystals

- Cholesterics

Locally a cholesteric (Ch) (Figure 1.3) is very similar to a nematic with a long range orientational order but because of the presence of chirality this nematic director \mathbf{n} twists spontaneously in a direction perpendicular to itself. This results in a helical structure with the director confined everywhere perpendicular to the helical axis. If the helical axis is taken to be along the z axis, then the cholesteric can be described as follows:

$$n_x = \cos(q_0 z + \phi)$$

$$n_y = \sin(q_0 z + \phi)$$

$$n_z = 0$$

Here the initial phase ϕ of the helix is arbitrary. Because of the equivalence of \mathbf{n} and $-\mathbf{n}$ the spatial period is equal to one half of the pitch $P = 2\pi/q_0$.

- Chiral Smectics

A chiral smectic or Smectic C^* (S_C^*) is a smectic C spontaneously twisted about the layer normal. Because of the helical arrangement of the molecules forming the phase, the symmetry of this mesophase is C_2 with a two-fold axis perpendicular to the twist axis. Even an electric polarisation is allowed along the two-fold axis. This phase is known as ferroelectrics. In general, in all these phases, in addition to the 1-d density wave, the director (Figure 1.3) \mathbf{n} which is at a constant tilt angle θ with the layer normal precesses about the layer normal from layer to layer. That is, the vector \mathbf{c} rotates with a periodicity P , the pitch of the structure.

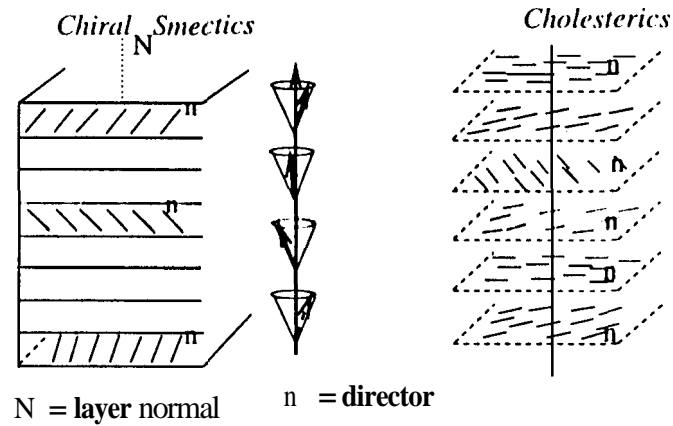


Figure 1.3: *Schematic* representation of chiral *smectics* and *cholesterics*

- Twist Grain Boundary Smectics

Many S_A with chiral molecules melt on heating, into a cholesteric phase. But recently, [2], [3, 4] an intermediate phase transition has been seen in some of these. For example between S_A and cholesterics one finds a new phase called the Twist grain boundary smectic. Essentially, it is a helical stack of thick S_A domains connected by thin grain boundaries made up of an array of screw dislocations. Inside each such boundary, the director n twists through an appropriate amount to smoothly connect the adjacent smectic blocks. This is schematically shown in Figure 1.4. Such structures are also possible near a S_A - S_C phase boundary. Hence between S_A and S_C we find a twist grain boundary smectic with S_C blocks connected by grain boundaries.

- Blue Phases

In general, on heating, cholesteric melts into an isotropic phase. But new thermodynamically stable phases appear between the cholesteric and isotropic phase on heating cholesterics of extremely small pitch. They

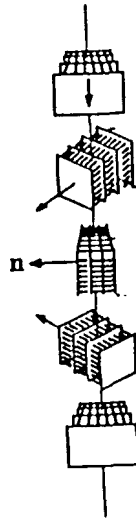


Figure 1.4: Schematic representation of a twist grain boundary smectics

are called as Blue phases. Structurally, most of these phases are a three dimensional lattice made of disclination lines.

Ferro liquid crystals

Liquid crystals can be doped with ferromagnetic grains with a very low grain concentration. One can prepare the system such that these grains align with their magnetization everywhere along the local director n i.e., the medium has a magnetisation M . In recent times, such systems have been made in the laboratory. For example, in the case of nematics, the liquid crystal is doped by taking it to the isotropic phase in the presence of an external magnetic field and then gradually cooling it to the nematic phase. The resultant phase is a ferronematic phase [5, 6]. Ferrocholesterics and ferrosmeectics have also been realised [7, 8].

1.2 Topological defects-Solitons

A large variety of topological defects exist in liquid crystalline phases. These defects are associated with elastic distortions in the director field and in some cases the elastic distortions of the layers. Topological defects in any system are decided by its inherent symmetry. Unlike the case of crystals where energetically favoured topological defects are dislocations of translation, in liquid crystals the most common topological defects are dislocations of rotation called disclinations. Since liquid crystal phases are made up of highly anisotropic molecules some of the physical properties are also anisotropic in nature. For instance, liquid crystals have anisotropy in their dielectric and diamagnetic susceptibilities. Generally, the diamagnetic anisotropy (χ_a) is positive for liquid crystals made up of rod-like molecules and negative for liquid crystals made up of disc-like molecules. Due to this, naturally existing defects get modified in the presence of an external magnetic field. Also, there are some defect structures which are new and which are natural topological objects in an external field. One such a defect is a *Wall* or a *Planar Soliton*. As this thesis largely deals with planar solitons we describe them briefly here.

1.2.1 Static Planar Solitons

A planar soliton, as said earlier is a natural topological object in the presence of an external magnetic field. The structure of such a wall in nematics was first studied by Helfrich [9]. These walls are similar to the Bloch and Neel walls present in ferro-magnetic systems. In a nematics, because of the 180° rotational symmetry perpendicular to the director, both n and $-n$ have the

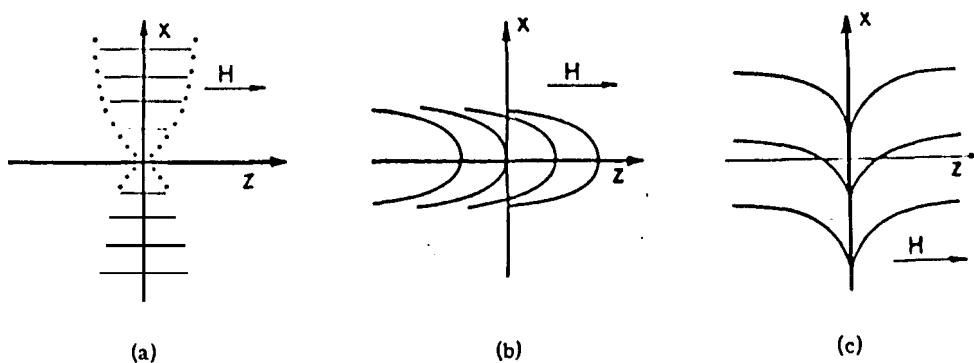


Figure 1.5: Helfrich Walls. (a) Twist soliton (b) Bend-rich soliton (c) Splay-rich soliton

same magnetic potential energy. This magnetic potential energy is minimum, for a $\chi_a > 0$ nematics, whenever \mathbf{n} is parallel or **anti-parallel** to \mathbf{H} . Thus in the presence of a field these two states which are **energetically** degenerate can be smoothly connected by a Wall or a *soliton* [10] [11]. The orientation of the director continuously changes by π on crossing the wall. For this reason, they can also be called π solitons. We can have twist solitons, bend-rich solitons and splay-rich solitons as depicted in Figure 1.5. These structures are strictly non-singular and most of the distortion is confined to a narrow region. These solitons are different from "true" solitons which are traveling solitary waves and which retain their shape and structure after **pairwise** collision. For the sake of simplicity we briefly discuss here a *Twist Soliton*.

Let the magnetic field \mathbf{H} be along the z axis and the director is confined to the $y - z$ plane with variations along the x axis i.e., $\mathbf{n} = (0, \sin \phi, \cos \phi)$ with $\phi = \phi(x)$. The free energy density of distortion of nematics in an external

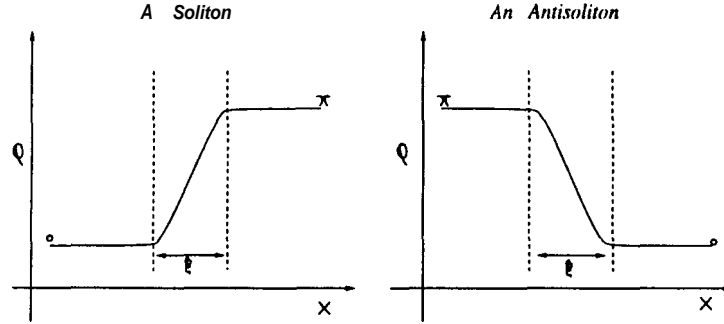


Figure 1.6: Profile of a soliton and an antisoliton

magnetic field is given by:

$$F = F_{elastic} - \frac{\chi_a}{2}(\mathbf{n} \cdot \mathbf{H})^2 \quad (1.1)$$

where $\chi_a = \chi_{||} - \chi_{\perp}$. $\chi_{||}$ and χ_{\perp} are diamagnetic susceptibilities parallel and perpendicular to \mathbf{n} .

$$F_{elastic} = \frac{K_{11}}{2}(\nabla \cdot \mathbf{n})^2 + \frac{K_{22}}{2}(\mathbf{n} \cdot (\nabla \times \mathbf{n}))^2 + \frac{K_{33}}{2}(\mathbf{n} \times (\nabla \times \mathbf{n}))^2 \quad (1.2)$$

$F_{elastic}$ is the free energy density for distortions in the director field [12]. Here $\nabla \cdot \mathbf{n}$ represents the splay distortion, $\mathbf{n} \cdot (\nabla \times \mathbf{n})$ represents the twist distortion and $\mathbf{n} \times (\nabla \times \mathbf{n})$ represents the bend distortion. The coefficients K_{11} , K_{22} and K_{33} are the respective elastic constants. In this particular case, only pure twist is present and the minimisation of total energy gives,

$$\frac{\partial^2 \phi}{\partial x^2} = \phi_{xx} = \frac{\chi_a H^2}{K_{22}} \sin \phi \cos \phi \quad (1.3)$$

where ϕ is the angle between the field and the director. This can be solved, subject to boundary conditions $\phi(x = -\infty) = 0$, $\phi(x = +\infty) = \pi$ and $\partial\phi/\partial x = 0$

at $x = \pm\infty$. The equation (1.3) has a solution given by

$$\phi = 2 \tan^{-1} (e^{x/\xi}) \quad (1.4)$$

It shows that the twist distortion is confined to a narrow length called the coherence length ξ with

$$\xi = \frac{1}{H} \sqrt{\frac{K_{22}}{\chi_a}} \quad (1.5)$$

Figure 1.6(a) gives the ϕ profile of a soliton. The total energy per unit area of the wall is given by

$$E = \int_{-\infty}^{+\infty} F dx = 2H \sqrt{\chi_a K_{22}} \quad (1.6)$$

So even though the wall is of infinite extent its energy is finite. Equation (1.3) also permits the solution

$$\phi = 2 \tan^{-1} (e^{-x/\xi}) \quad (1.7)$$

This represents an antisoliton with $\phi(x = -\infty) = \pi$, $\phi(x = +\infty) = 0$. Its profile is also shown in Figure 1.6(b). Similar results are found for the case of splay-rich and bend-rich walls.

In this thesis, we are primarily concerned with the mechanical and optical properties of such soliton states. While undertaking these studies we found some new planar soliton states in nematics. A brief discussion on these solitons is presented here.

1.3 Some new soliton states in nematics

1.3.1 Intertwined Solitons

In nematics with negative diamagnetic anisotropy ($\chi_a < 0$), in a magnetic field H , a topologically permitted non-singular defect solution is a cylindrically

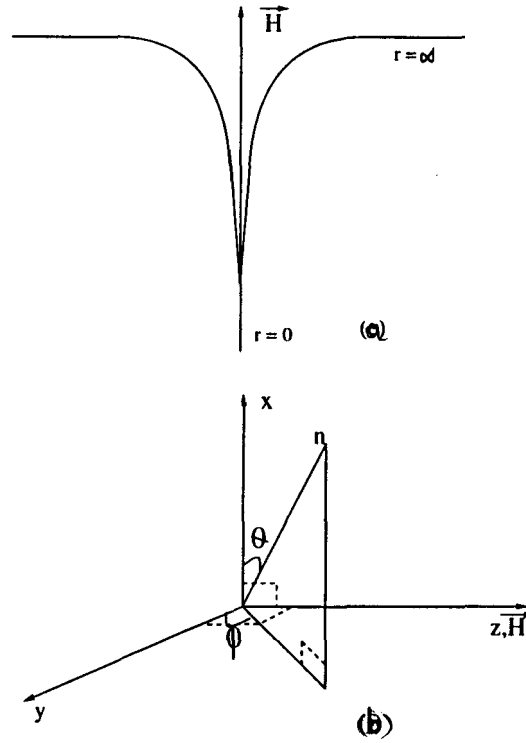


Figure 1.7: (a) **All** radial splay-bend soliton. (b) The coordinate system **with** reference to which the planar solitons shown in Figure 1.8 are described.

symmetric structure such that at $r = \infty$, the director \mathbf{n} is perpendicular to \mathbf{H} and is along \mathbf{H} at $r = 0$. It may be mentioned that \mathbf{n} can be either in an all radial or all circular state at $r = \infty$. Figure (1.7) shows an all radial structure. This is also a soliton. We can also construct planar solitons in such nematics. Here also there are three possible planar solitons viz., the twist soliton, the bend soliton and the splay soliton. These are similar to planar solitons of $\chi_a > 0$ nematics. Here we refer \mathbf{n} and \mathbf{H} to a coordinate system shown in Figure 1.7.

In a bend soliton (Figure 1.8(a)) described by $\mathbf{n} = (0, \cos \phi, \sin \phi)$ with

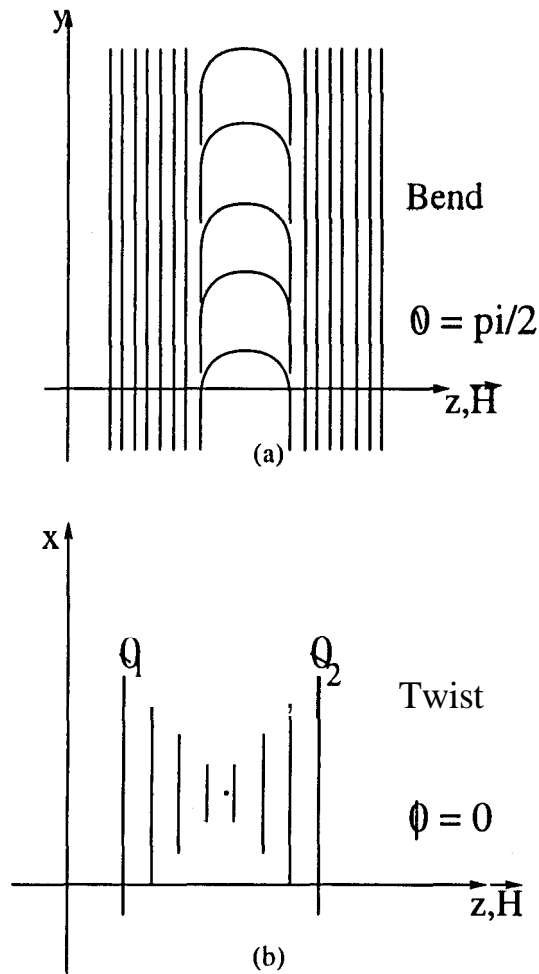


Figure 1.8: *Planar Solitons. (a) π bend soliton confined to the $y - z$ plane with (b) A pure twist deformation about z*

$\phi = 2 \tan^{-1} e^{z/\xi}$ the director is perpendicular to z axis at $z = \pm\infty$ and $n_x = 0$ everywhere. These states are of minimum energy since $\chi_a < 0$ and they get connected by a continuous bend in the director in the $y - z$ plane.

We now consider the effect an out of plane distortion in this well known planar solitons. That is, we impose a twist distortion θ on n . The director

now has components $n_x = \cos \theta$, $n_y = \sin \theta \cos \phi$ and $n_z = \sin \theta \sin \phi$. The free energy density in the one constant approximation i.e., $K_{11} = K_{22} = K_{33} = K$

$$F = \frac{K}{2} [(\nabla \theta)^2 + (\sin \theta)^2 ((\nabla \phi)^2 - E_2 (\sin \phi)^2)] \quad (1.8)$$

Here $E_2 = \chi_a H^2 / K$. The equations of equilibrium are

$$\nabla^2 \theta = \sin \theta \cos \theta [(\nabla \phi)^2 - E_2 (\sin \phi)^2] \quad (1.9)$$

$$\nabla^2 \phi = -E_2 \sin \phi \cos \phi - \cot \theta [2(\nabla \theta \cdot \nabla \phi)] \quad (1.10)$$

We seek bend soliton solution for ϕ assuming that the θ variation is also along the direction of ϕ variation i.e., along z . Then we can see that the out of plane distortion can only be a twist distortion connecting the same base states of the bend soliton. From Figure 1.8(b) we see that a pure 9 distortion confines the director everywhere to a plane perpendicular to the field and since we are considering $\chi_a < 0$ nematics, its elastic distortion is not coupled to the field. Hence left to itself it will unwind on its own and go to the uniform state. But interestingly in view of (1.9) and (1.10) we find that this twist in θ can be coupled to the field through variations in ϕ . The numerical solutions of these equations which reveal this aspect is shown in Figure 1.9 (a) to (d). Interestingly, even the 8 variations are confined to a narrow region of space as in a soliton. It has a net twist $\Delta \theta = \theta_1 - \theta_2$. Hence even the out of plane distortion is in the form of a soliton. The value of the net twist $\Delta \theta$ is a function of H . It can be made to change continuously from 0 to π . In Figure 1.9(d) we see that a π bend soliton is coupled with a π twist soliton. We call this an intertwined soliton. The twist per unit length (as given by the slope at the centre) in the

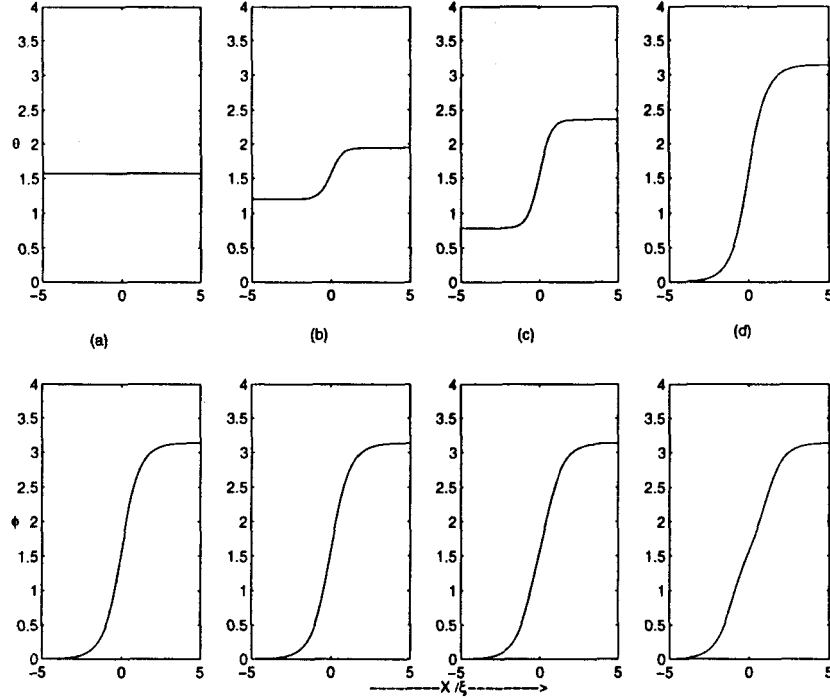


Figure 1.9: The amount of twist locked in θ is shown with the corresponding π soliton in χ .

$\Delta\theta$ soliton is seen to increase with increasing **Ad**.

Similarly, we can construct two other types of intertwined solitons. 1) A twist soliton with twist along x axis intertwined with a splay soliton with variation along the y axis. 2) A splay soliton with variation along x axis intertwined with a bend soliton with variation along y axis.

Interestingly we find such intertwined soliton states only in $\chi_a < 0$ nematic. We can easily understand this because a θ distortion in the $\chi_a > 0$ nematics leads to an increase in the magnetic energy.

1.3.2 Soliton-antisoliton pairs

The solitons described in the earlier sections are static. However we can also have dynamic solitons, Dynamic solitons result from one of the following two qualitatively different kinds of mechanisms.

- Motion due to base state asymmetry

If it so happens that a soliton connects director orientations of different magnetic potential energies, then it will move, with the stable state expanding into the unstable region.

- Motion due to core asymmetry

It may also happen that even though a soliton connects base states of the same magnetic potential energy, the symmetry in the magnetic potential energy can be broken inside the soliton. This also leads to an expansion of the stable region into the unstable region and hence to a motion of the soliton.

A soliton in a nematic confined to a plane and subjected to a rotating magnetic field about an axis perpendicular to the plane, can move due to core asymmetry. We discuss such a soliton here.

Dynamic solitons in nematics

We consider a nematic with the director \mathbf{n} confined to a plane, say the xy plane with the magnetic field \mathbf{H} also acting in the same plane but rotating with an angular frequency ω about the z axis. The differential equation for the director field of a nematic in this geometry is given by [13]

$$K \frac{\partial^2 \phi}{\partial x^2} = \gamma_1 \frac{\partial \phi}{\partial t} + \frac{\chi_a}{2} H^2 \sin 2\phi - \gamma_1 \omega \quad (1.11)$$

Here ϕ is the angle between \mathbf{n} and \mathbf{H} , K is the bend or splay elastic constant in the one constant approximation and γ_1 is the rotational viscosity. This equation permits the solution

$$\sin 2\phi_0 = \frac{2\gamma_1\omega}{\chi_a H^2} = \frac{\omega}{\omega_c}$$

where $\omega_c = \frac{\chi_a H^2}{2\gamma_1}$

For $\omega < \omega_c$, this represents a uniform director field co-rotating with the field \mathbf{H} at a constant phase lag angle ϕ_0 . This is called the synchronous regime. For $\omega > \omega_c$, the angle ϕ_0 is not given by the above relation. It can be shown that in this region the angle ϕ_0 between \mathbf{n} and \mathbf{H} becomes a function of time. This is the asynchronous regime.

In the uniform director field \mathbf{n} of the synchronous regime (i.e., $\omega < \omega_c$) we can construct a planar soliton. It can either be a bend-rich or a splay-rich soliton with the director confined to the $x - y$ plane. In these solitons, the uniform states will be at an angle ϕ_0 to the field \mathbf{H} . We have shown in Figure 1.10 a splay-rich soliton and an antisoliton. It can be seen from the figure that the uniform states are at the same angle ϕ_0 with respect to \mathbf{H} while the director field inside the wall has one region more favourably aligned with respect to \mathbf{H} . Thus the soliton moves. We hence seek traveling wave soliton solutions of (1.11). In a coordinate system $\mathbf{X} = x - ut$, moving with a velocity u , equation (1.11) simplifies to,

$$\phi_{XX} = \eta\phi_X + \beta \sin 2\phi - F \quad (1.12)$$

where $\beta = \frac{\chi_a H^2}{2K}$, $F = \gamma_1\omega/K$ and $\eta = \gamma_1 u/K$. In all our computations we have taken $K = 10^{-6}$ dynes, $\chi_a = 10^{-6}$ cgs units and $\gamma_1 = -0.53$ poise. This differential equation permits a traveling wave soliton solution going from ϕ_0

Soliton-antisoliton pair in nematics in a rotating magnetic field

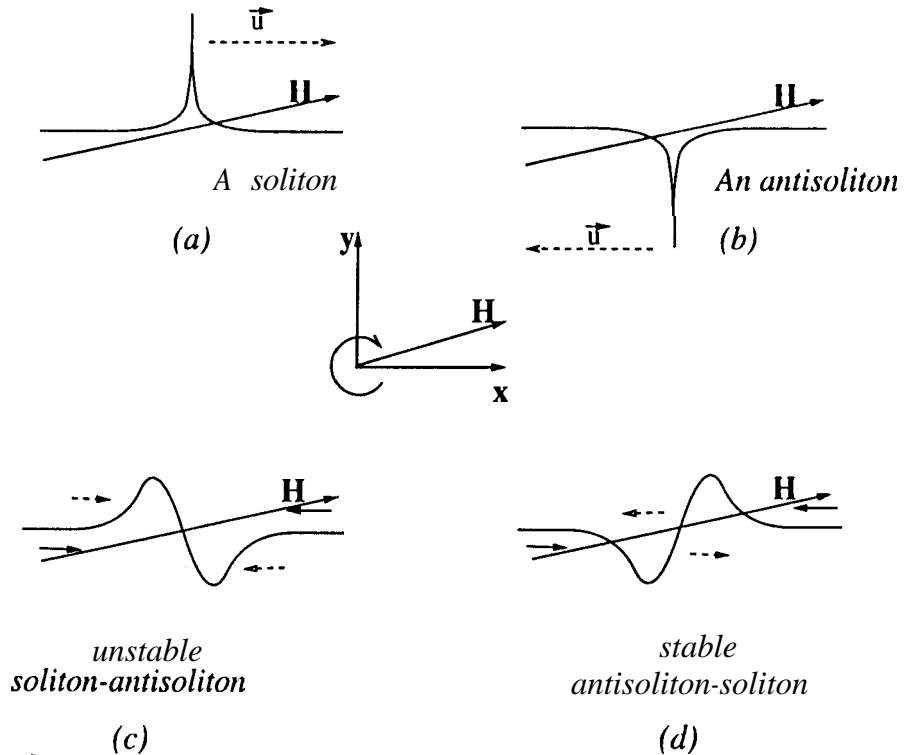


Figure 1.10: Schematic director representation of (a) A soliton, (b) Antisoliton (c) an unstable soliton-antisoliton pair (d) stable antisoliton-soliton pair in nematic. The full arrow and the dotted arrow represent the direction of force due to elastic and dynamic interactions respectively between soliton pairs.

to $\phi_0 + \pi$ traveling at a velocity u in a direction perpendicular to the wall. It can be shown that u , the velocity of propagation of the soliton increases monotonically from 0 at $F = 0$ and diverges sharply at $F = \beta$. In the same way we find that, the antisoliton moves in a direction opposite to the soliton i.e., its velocity is $-u$. That is, due to the core asymmetry if the soliton moves in $+x$ direction the antisoliton moves in $-x$ direction.

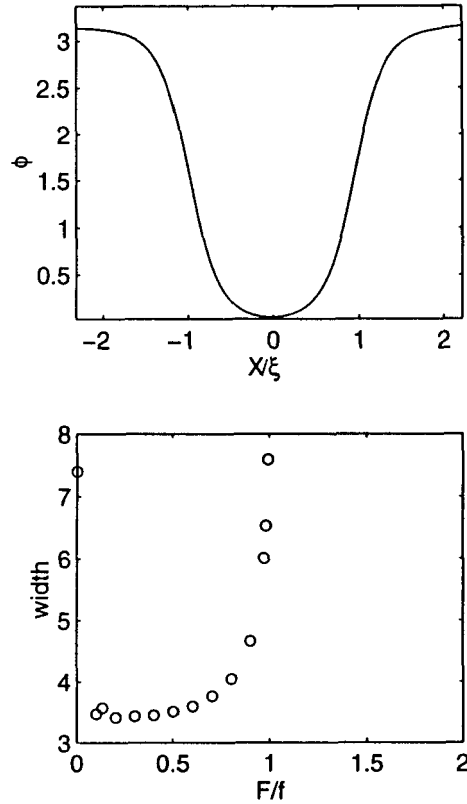


Figure 1.11: (a) Profile of antisoliton-soliton pair in nematics. (b) The variation of *width* (which has been scaled relative to ξ) of the pair with F/f

Static soliton-antisoliton pairs

In a static magnetic field, if a soliton and an antisoliton simultaneously exist they will attract and annihilate one another leading to an uniform state. However in a dynamical situation as in a rotating magnetic field, a soliton and an antisoliton can coexist with a finite separation. This however, happens only in situations where the opposing motion imparted by the core asymmetry opposes the attractive interaction between these unlike soliton pairs.

For example, in the present problem, an antisoliton-soliton pair (Figure 1.10(d)) is stabilised while a soliton-antisoliton pair (Figure 1.10(c)) is unsta-

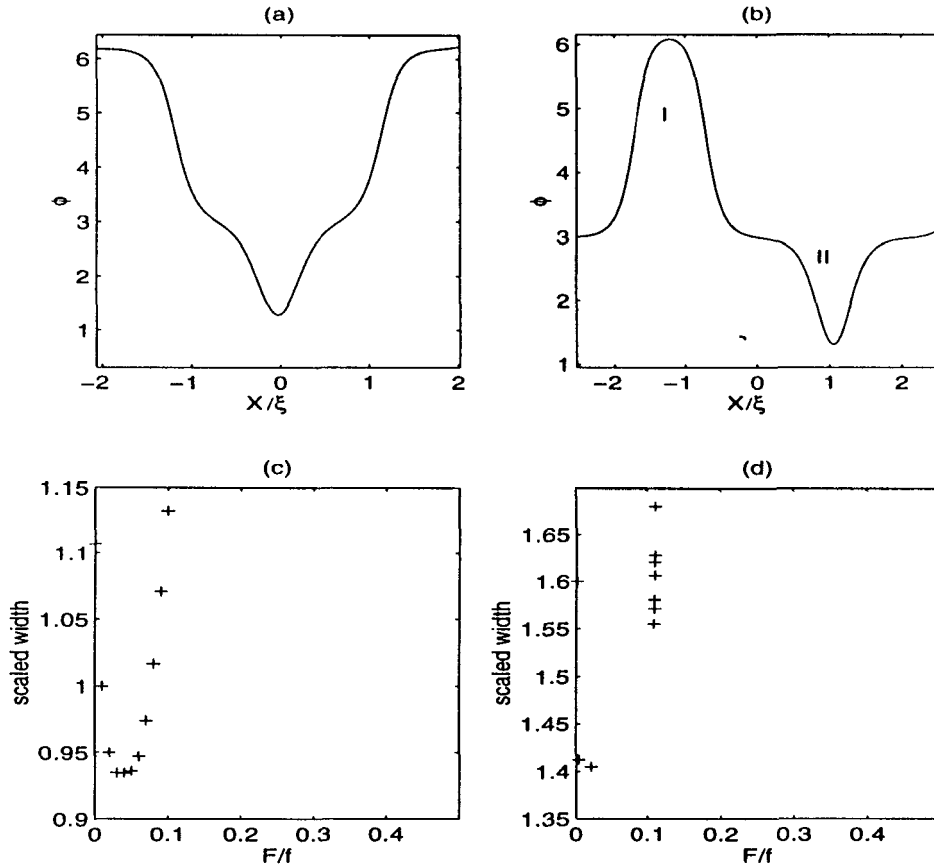


Figure 1.12: Profiles of unlike soliton pairs in a ferronematic. (a) a static antisoliton-soliton pair (b) a static soliton-antisoliton pair bound to an antisoliton-soliton pair (I). ϕ is in radians. Widths of unlike soliton pairs as a function of F/f . (c) for soliton structure shown in (a), (d) for the structure shown in (b). Widths have been scaled relative to ξ . $M = 0.001$ and $\chi_a = 10^{-6}$

ble. Profile of antisoliton-soliton structure in nematics is shown in Figure 1.11. The width defined as the separation at half maximum is a sensitive function of F . It is important to note that the width of antisoliton-soliton structure diverges both as $F \rightarrow 0$ and as $F \rightarrow f = \beta$. The variation of width with F/f is shown in Figure 1.11(b). This can be understood as follows. When $F = 0$, the permitted state is a static single soliton and when $F = f$ the velocities of the soliton and antisoliton diverge. This leads to a divergence in the width at both $F = 0$ and $F = f$.

The above arguments for a nematic can also be extended to a ferronematics - a nematic doped with magnetic grains. Unlike nematics which has only two stable states at $\phi = 0$ and $\phi = \pi$ ferronematics has two stable states at $\phi = 0$ and $\phi = 2\pi$. That is, the natural topological object is a 2π soliton and the dynamical equation (1.12) has an additional $MH \sin \phi$ term. Depending upon the strengths of various terms contributing to the magnetic potential energy, this system, in some parameter regimes, can have a metastable state at $\phi = \pi$ as well.

New kinds of soliton-antisoliton pairs can exist in this system. The detailed structure is decided by the sign of χ_a . Here we give the structure for $\chi_a > 0$ ferronematics. Figures 1.12(b) and (c) show two such soliton-antisoliton pairs. Their associated divergence in their widths are also shown in the same figure. The unlike pair shown in Figure 1.12(c) is rather similar to that found in a nematic excepting for a finer structure. When $M > \chi_a H$ this will not have this fine structure and the profile is then very similar to that found in a nematic. However the bound state shown in Figure 1.12(b) is unique to ferronematics. Note that, the widths of these structures diverges at $F = 0$ and at $F < f$. The divergence at $F < f$ is unlike the nematic case where it takes place for $F = f$. Here

$$f = \beta \sin 2\phi_m + \alpha \sin \phi_m \quad (1.13)$$

with $\alpha = MH/K$. Here ϕ_m defines the maximum possible angle between \mathbf{n} and \mathbf{H} in the synchronous regime.

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