Field Induced Chiral-Achiral Transitions in Liquid Crystals

N. Andal and G. S. Ranganath (*)

Raman Research Institute, Bangalore-560080, India

(Received 10 July 1995, revised 1 December 1995, accepted 8 February 1996)

PACS.61.30.Jf – Defects in liquid crystals
PACS.64.70.Md – Transitions in liquid crystals

Abstract. — We have considered chiral-achiral phase transitions that can occur in some liquid crystals. This study deals with such transformations induced by external electric and magnetic fields. We discuss phase changes mediated by disclinations, single solitons, soliton lattices and those occurring without any defect. Some unusual aspects of these transitions such as reentrance, occurrence of tricritical points and Lifshitz points have been highlighted.

1. Introduction

Phase transitions induced in liquid crystals by external electric and magnetic fields [1] is an important area of work since they have many unusual aspects associated with them. In this paper we study field induced chiral-achiral phase transitions occurring in some liquid crystal systems. Here the term *chirality* refers to a structural twist occurring on a macroscopic scale. This is a manifestation of the intrinsic chirality of either the constituent molecules or the dopents. Structural transformations from a twisted configuration to an untwisted configuration or *vice-versa* have been studied. They can occur with or without the mediation of topological defects. In the defect mediated processes we have considered transitions triggered by disclinations, single solitons and soliton lattices. We have paid attention to the cholesteric-nematic, ferrocholesteric (FCh)-ferronematic (FN), and chiral-achiral transitions in ferrosmectics (FS) systems. We find many interesting features in these phase transformations leading to rich phase diagrams with tricritical points, Lifshitz points and the phenomena of reentrance.

2. Transitions with the Magnetic Field along the Axis of Symmetry

2.1. FERROCHOLESTERICS. — A ferrocholesteric (FCh) is obtained from a usual cholesteric by doping it with magnetic grains so that the local magnetization **M** is along the local director n i.e., **M** spirals uniformly about the twist axis with a pitch P. When unwound such a system would become a ferronematic (FN). Such systems have been realized in the laboratory [2], with a very good mechanical coupling between **M** and **n**. The transition of an FCh to an FN in the presence of a magnetic field applied perpendicular to the twist axis is already well investigated [3, 4]. Here we consider the FCh-FN transition in a field applied parallel to the twist axis of an FCh.

(*) Author for correspondence (e-mail: gsr@rri.ernet.in)

2.1.1. Ferrocholesteric to Ferronematic Transition. — The unperturbed state of the FCh is described by $\mathbf{n} = (\cos\phi_0, \sin\phi_0, 0)$ with $\phi_0 = 2\pi z/P$. The applied field is in the z direction. It results in an out of plane distortion in the director $\mathbf{n}$ described by $n_x = \sin\theta\cos\phi$, $n_y = \sin\theta\sin\phi$ and $n_z = \cos\theta$. The free energy density is given by

$$F = \frac{K}{2}[(\theta_z)^2 + \sin^2\theta(\phi_z^2 - 2q_0\phi_z)] - \frac{\chi_a H^2}{2}\cos^2\theta - MH\cos\theta \tag{1}$$

Here $\theta_z = \partial\theta/\partial z$ and $\phi_z = \partial\phi/\partial z$, χ_a is the diamagnetic anisotropy, $q_0 = 2\pi/P$ and K is the elastic constant in the one constant approximation. Minimization of the total energy yields

$$\theta_{zz} = \sin\theta\cos\theta[(\phi_z)^2 - 2q_0\phi_z + f] + g\sin\theta \tag{2}$$

$$\sin^2\theta\phi_{zz} = -2\sin\theta\cos\theta[\theta_z(\phi_z - q_0)] \tag{3}$$

where $f = \chi_a H^2/K$, $g = MH/K$, $\phi_{zz} = \partial^2\phi/\partial z^2$ and $\theta_{zz} = \partial^2\theta/\partial z^2$.

The equations (2) and (3) permit the following solutions:

$$\phi = \phi_0 = q_0 z \tag{4}$$

$$\cos\theta = \frac{MH}{Kq_0^2 - \chi_a H^2} \tag{5}$$

It is clear from equations (4) and (5) that in the presence of the external magnetic field the pitch of the structure is unaltered and that θ is uniform throughout. When $0 < \theta < \pi/2$ we get a tilted FCh. Since $\theta = 0$ in an FN we find that the transition from an FCh to an FN occurs when $MH = Kq_0^2 - \chi_a H^2$. The angle θ continuously decreases from $\pi/2$ as H increases from zero.

2.1.2. Ferronematic to Ferrocholesteric Transition. — In this section we study the transition from an FN to an FCh. We consider an FN obtained from an FCh by the application of a magnetic field along its twist axis. In the nematic state the director lies along the z direction. As we lower the field θ and ϕ distortions set in described by $n_x = \sin\theta\cos\phi$, $n_y = \sin\theta\sin\phi$ and $n_z = \cos\theta$. Below a particular value of H given by the solution of

$$\chi_a H^2 + MH - Kq_0^2 = 0 \tag{6}$$

a tilt θ, in the director $\mathbf{n}$, away from the field $\mathbf{H}$ develops. In view of the degeneracy in θ with respect to the field direction. we set up the equations of equilibrium in cylindrical polars (r, α, z). This leads to

$$\phi = q_0 z \pm N\alpha, N = \text{integer} \tag{7}$$

and the θ distortion obeys the differential equation

$$\theta_{rr} + \frac{1}{r}\theta_r = \frac{\sin\theta\cos\theta}{r^2} + (f - q_0^2)\sin\theta\cos\theta + g\sin\theta \tag{8}$$

Equations (7) and (8) permit a non-singular topological defect in $\mathbf{n}$. At the centre of this defect θ is zero and far away from it θ is given by (5). This solution is very similar to the N-flower solution considered in reference [5] for a $\chi_a < 0$ FN. In the present problem a similar topological defect arises though $\mathbf{M}$ is parallel to $\mathbf{H}$ and $\chi_a > 0$. Here the director at $r = \pm\infty$ is at a constant angle θ_0 with respect to the field and at the centre of the defect the director is along the field direction. Through the formation of such topological defects we can enter the tilted FCh state. Given enough time, the unlike defects will attract and annihilate one

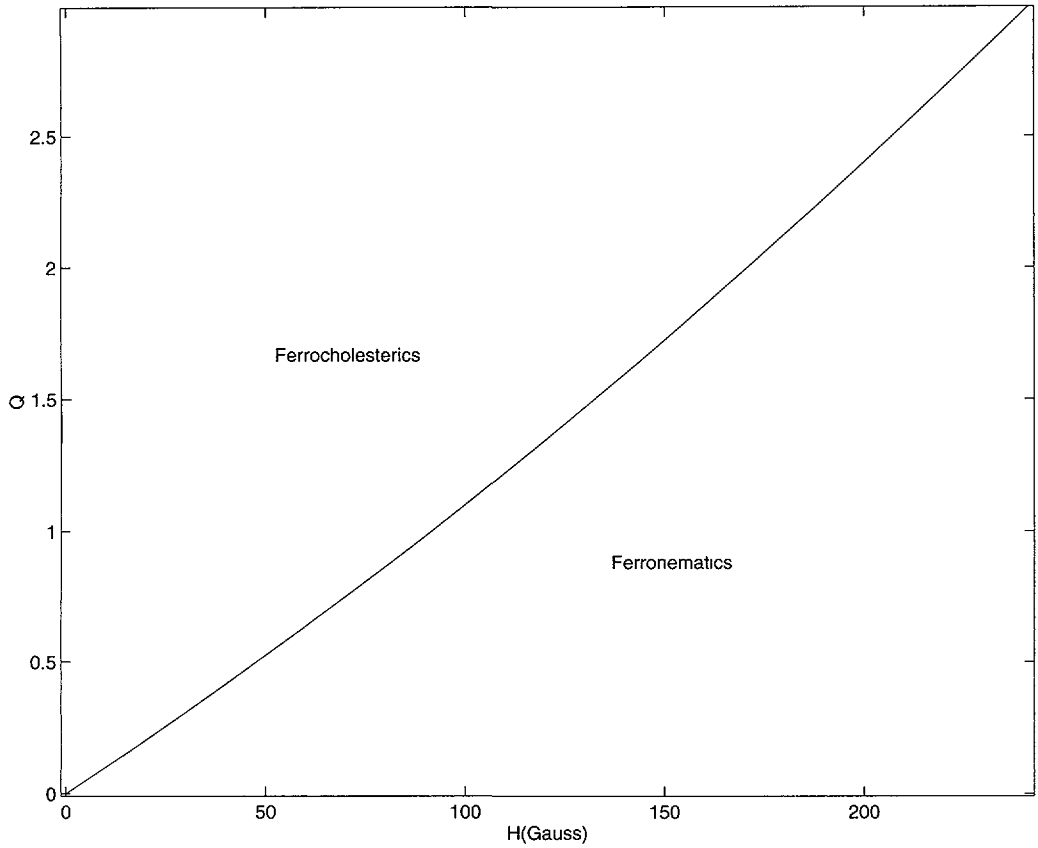

Fig. 1. — The transition from an FCh to an FN when $\chi_a > 0$. $Q = q_0^2$ in units of 10^{-5} cm^{-2}. $q_0 = 2\pi/P$ with P as the pitch of the FCh and H is the magnetic field. $M = 0.0001$ Gauss and $\chi_a = 10^{-6}$ cgs units.

another leading to an uniformly tilted FCh. Hence in this system one possible structural transformation, which is permitted by the equations of equilibrium, is that the achiral to chiral transition can be defect mediated while the chiral to achiral can take place without defects.

It must be pointed out that in the case of FN $\rightarrow$ FCh transition, we can also have uniform θ solution without the formation of defects provided there is a predisposition of the director to tilt in a particular direction due to sample boundaries.

2.1.3. Phase Diagrams. — In FCh, for $\chi_a > 0$, on a gradual increase of the magnetic field, θ decreases monotonically and at a critical field H_c, FCh goes over to the FN state. The phase diagram for this transition is shown in Figure 1. Here we have presented the phase diagram in the H, $Q(= q_0^2)$ space since it is possible to have cholesteric systems where q_0 can be varied continuously. This is possible both in compensated cholesterics [6] and in some pure systems [7]. We find a totally different phase diagram when $\chi_a < 0$. This is shown in Figure 2. We see that depending on the value of q_0^2 we get a reentrance of FCh on increase of the external field. For q_0^2 above a critical value the transition from FCh to FN does not take place at all. This phase diagram can be easily understood from the fact that there are two opposing torques on the director, one due to the χ_a term which tilts the director towards the cholesteric

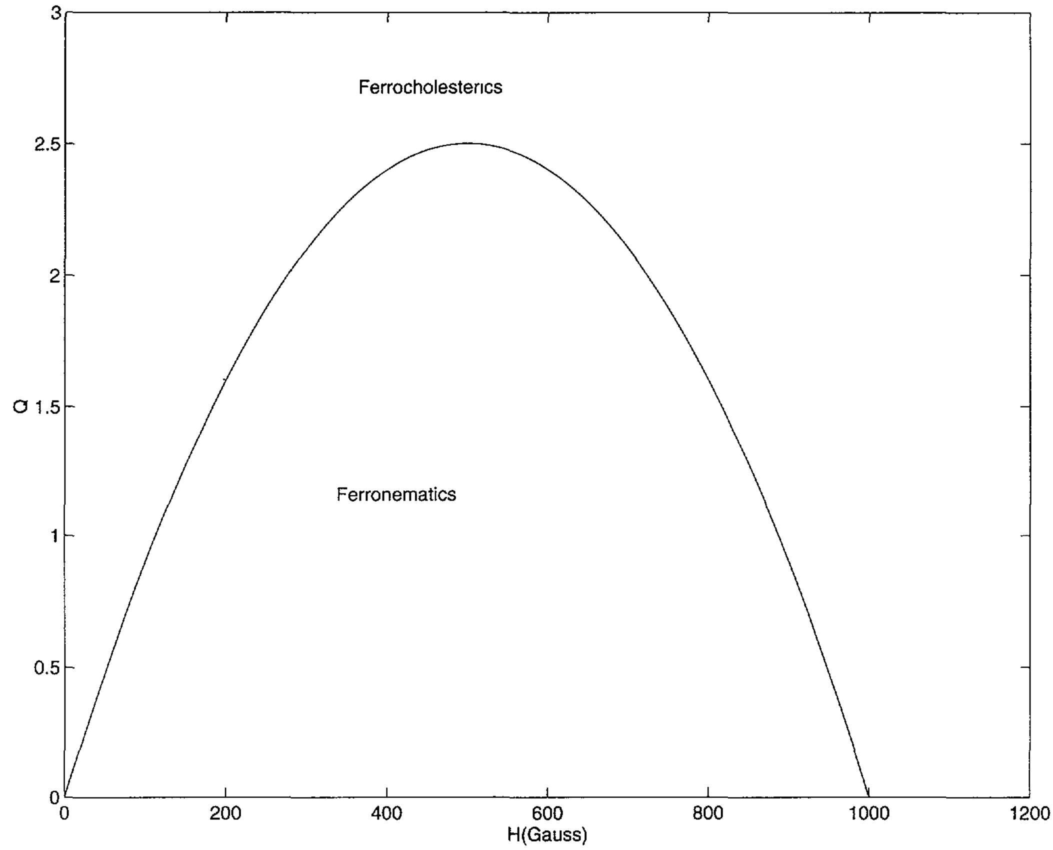

Fig. 2. — The transition from an FCh to an FN when $\chi_a < 0$. $M = 0.0001$ Gauss and $\chi_a = -10^{-6}$ cgs units.

planes and the other due to the M term which tilts it towards the twist axis. The net torque decides the state of the system. In view of the discussions presented in Section 2.1.2 we expect the FN to FCh transition to be mediated by N-flower defects.

2.2. Ferrosmectics. — Ferrosmectics (FS) are smectic systems which have been doped with magnetic grains. Such systems have already been made in lyotropic liquid crystals [8, 9]. We consider here an FS with the magnetic grains aligned such that the local magnetization $\mathbf{M}$ is parallel to the local director $\mathbf{n}$. We further make the system chiral by doping it with chiral molecules. It is possible for such a system to have a low temperature chiral smectic C* like phase ($\mathrm{FS_{C^*}}$) and a high temperature achiral phase of smectic C ($\mathrm{FS_C}$) or smectic A ($\mathrm{FS_A}$) type. In our analysis of this system we take the electric polarization in the chiral phase to be negligible. We have considered the chiral-achiral transitions near the $\mathrm{FS_C(FS_A)}$-$\mathrm{FS_{C^*}}$ point. Here the tilt θ of $\mathbf{n}$ with respect to the layer normal can be assumed to be small.

2.2.1. Ferrosmectic C to Ferrosmectic A Transition.* — We first discuss the transition from $\mathrm{FS_{C^*}}$ to $\mathrm{FS_A}$. The smectic layers of $\mathrm{FS_{C^*}}$ are in the $x-y$ plane with the director at an angle θ_0 with the layer normal. In the absence of the field the director configuration is $\mathbf{n_0} \approx (\theta_0 \cos\phi_0, \theta_0 \sin\phi_0, 1)$ with $\phi_0 = 2\pi z/P$, P being the pitch of the helical structure. In the presence of a magnetic field **H** along z we find $n_x \approx \theta\cos\phi$, $n_y \approx \theta\sin\phi$ and $n_z \approx 1$. The free energy density is,

$$F = \frac{\alpha}{2}\theta^2 + \frac{\beta}{4}\theta^4 + \frac{K}{2}[(\nabla\theta)^2 + \theta^2(\phi_z^2 - 2q_0\phi_z)] + \frac{1}{2}\chi_a H^2\theta^2 \pm \frac{MH\theta^2}{2} \tag{9}$$

The parameters α and β are the Landau coefficients, χ_a is the diamagnetic anisotropy which is assumed to be positive, K is the elastic constant for ϕ distortions in the one constant approximation and $q_0 = 2\pi/P$. We consider the positive or the negative sign according as **H** is parallel or anti-parallel to $\mathbf{M}_z$, the component of **M** along to the twist axis.

In the FS_{C^*} phase α is negative equal to $-\alpha_0$. As in the case of ferrocholesterics here also in the chiral phase a uniform twist with a constant tilt are permitted solutions given by

$$\phi = \phi_0 = q_0 z \tag{10}$$

$$\theta = \sqrt{\frac{(\alpha_0 + Kq_0^2) - (\chi_a H^2 \pm MH)}{\beta}} \tag{11}$$

Transition from FS_{C^*} to FS_A occurs when

$$(\alpha_0 + Kq_0^2) - (\chi_a H^2 \pm MH) = 0 \tag{12}$$

This transition occurs by a continuous change in θ. In principle the critical fields for the transition are different for **H** parallel to $\mathbf{M}_z$ and anti-parallel to $\mathbf{M}_z$ cases. However with **H** anti-parallel to $\mathbf{M}_z$ and for $\chi_a < 0$ we do not get a transition to the FS_A state.

2.2.2. Ferrosmectic A to Ferrosmectic C Transition.* — Consider an FS_A in a magnetic field parallel to the layer normal. In the absence of the field we have $\mathbf{n}_0 = (0, 0, 1)$. In this geometry if the magnetization **M** is anti-parallel to **H**, we expect a tilt θ in the director **n** with a ϕ degeneracy in the plane of the smectic layers. The director components are $\mathbf{n} = (\theta\cos\phi, \theta\sin\phi, 1)$. Since the phase is assumed to lack a mirror symmetry due to the presence of chiral molecules, this tilted director **n** also precesses about the layer normal. In other words a tilt θ results in an azimuth ϕ which is a function of x, y and z. The free energy density for a $\chi_a > 0$ material is given by

$$F = \frac{\alpha}{2}\theta^2 + \frac{\beta}{4}\theta^4 + \frac{K}{2}[(\nabla\theta)^2 + \theta^2(\phi_z^2 - 2q_0\phi_z + (\phi_x^2 + \phi_y^2))] + \frac{1}{2}\chi_a H^2\theta^2 - \frac{MH\theta^2}{2} \tag{13}$$

In the present case $\alpha > 0$, as we are in the FS_A phase. Minimization of total energy leads to the following coupled equations.

$$\nabla^2\theta = \theta[a + (\phi_z)^2 - 2q_0\phi_z + (\phi_x^2 + \phi_y^2) + f - g] + b\theta^3 \tag{14}$$

$$\theta^2\nabla^2\phi = -2\theta[\nabla\theta\cdot(\nabla\phi - q_0\hat{\mathbf{k}})] \tag{15}$$

where $a = \alpha/K$, $b = \beta/K$ and $\hat{\mathbf{k}}$ is a unit vector along z. Equations (14) and (15) permit the following solutions in cylindrical polars:

$$\phi = q_0 z \pm N\alpha, \quad N = \text{integer}$$

And θ is assumed to be a function of r only. It obeys the differential equation

$$\theta_{rr} + \frac{1}{r}\theta_r = \theta[(a + (\frac{N}{r})^2 + f) - (q_0^2 + g)] + b\theta^3 \tag{16}$$

The solution in ϕ describes a disclination of strength N with its associated ϕ pattern rotating as we go along the z axis. Equation (16) is the familiar *Ginsburg-Piteavıskii* equation. The tilt angle θ goes from zero at the centre of the disclination to a constant value θ_0 at a large distance from the centre [10]. Such field induced disclinations start interacting soon after creation. Given enough time unlike disclinations will annihilate one another resulting finally in an uniformly twisted $\mathrm{FS_{C^*}}$ phase with a tilt angle θ_0. This θ_0 is given by

$$\theta_0 = \sqrt{\frac{(MH + Kq_0^2) - (\alpha + \chi_\mathrm{a} H^2)}{\beta}} \tag{17}$$

It is clear from (17), that only for certain values of H we get a transition to the $\mathrm{FS_{C^*}}$ (i.e., $\theta_0 \neq 0$) . It is to be noted that, in the case of $\chi_\mathrm{a} > 0$, for $\mathbf{H}$ parallel to $\mathbf{M}_z$ no phase transition to the $\mathrm{FS_{C^*}}$ takes place. And for $\chi_\mathrm{a} < 0$, both with $\mathbf{H}$ parallel and anti-parallel to $\mathbf{M}_z$ we find that a transition to $\mathrm{FS_{C^*}}$ is possible at a critical field H_c. Interestingly, the critical fields in the two cases are different. Thus we see that the above analysis permits a disclination triggered transition from $\mathrm{FS_A}$ to $\mathrm{FS_{C^*}}$ and a defect free transition from $\mathrm{FS_{C^*}}$ to $\mathrm{FS_A}$.

2.2.3. Phenomenon of Reentrance. — In the presence of the field, both in $\mathrm{FS_A}$ and $\mathrm{FS_{C^*}}$, we get the phenomena of reentrance. Figures 3 and 4 depict this. It should be noted that in both the cases the transition from the chiral to the achiral phase is not defect mediated while the transition from the achiral to the chiral phase is always through disclinations.

2.2.4. Tricritical Point. — By incorporating higher order terms in the magnetic field contribution to the free energy, we can show that at a certain field the coefficient of the θ^4 term can change sign. Beyond this field, the $\mathrm{FS_A} - \mathrm{FS_{C^*}}$ phase boundary becomes first order. Therefore we can expect a tricritical point on this phase boundary. Hence this phase change at high fields can become first order above a certain value of H while at low fields it is second order.

2.2.5. Grain Migration. — It is well known [3,4] that in a magnetic field acting perpendicular to the twist axis of an FCh the magnetic grains migrate out of highly distorted regions to regions of low distortion. The same phenomenon can be expected in $\mathrm{FS_{C^*}}$ also in the same geometry. We have seen that in both FCh and $\mathrm{FS_{C^*}}$ in a magnetic field parallel to the twist axis the ϕ and θ distortions are uniform all over. Hence in this geometry we find FCh to FN or $\mathrm{FS_{C^*}}$ to $\mathrm{FS_A}$ transition to take place without grain migration. Even when we go from FN to FCh or $\mathrm{FS_A}$ to $\mathrm{FS_{C^*}}$ though non-uniform distortions result due to the creation of defects, these get ironed out quickly due to the attraction between unlike defects. Therefore even here, there will be no grain migration. This is the unusual feature of these chiral-achiral transitions.

2.3. CHOLESTERIC-NEMATIC TRANSITION. — In this section we discuss the cholesteric (Ch)-nematic (N) transition. This transition, in a magnetic field perpendicular to the twist axis, takes place through the creation of a π soliton lattice [11,12]. The soliton lattice goes over to a nematic state at a critical field $H_\mathrm{c} = (\pi^2/P)\sqrt{K/\chi_\mathrm{a}}$. Here we consider Ch-N transformation in a magnetic field applied parallel to the twist axis. In this configuration the phase change can be effected through the creation of a single soliton. We consider only the case of χ_a positive materials since for a χ_a negative cholesteric. in this geometry, the field stabilizes the undistorted structure.

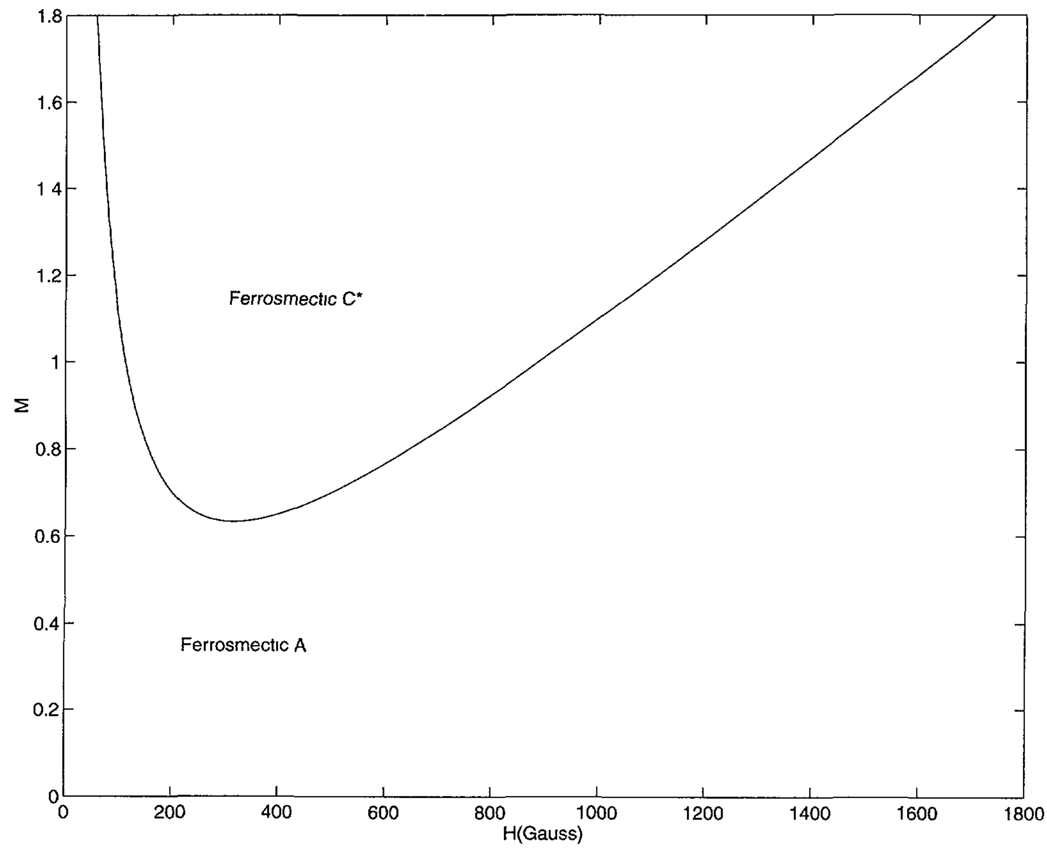

Fig. 3. — The phase diagram for the achiral FS_A phase. Here $\alpha = 1$, $\beta = 0.1$, $\chi_a = 0.1 \times 10^{-6}$ and $\mathbf{M}_z$ and $\mathbf{H}$ are anti-parallel to one another. $\mathbf{M}$ is in units of 10^4 Gauss.

2.3.1. Dechiralising Soliton in a Cholesteric. — The undistorted structure in the absence of the field is given by $\mathbf{n}_0 = (\cos\phi_0, \sin\phi_0, 0)$ with $\phi_0 = 2\pi z/P$. In a field parallel to the twist axis the director $\mathbf{n}$ will develop an out of plane distortion given by $n_x = \sin\theta\cos\phi$, $n_y = \sin\theta\sin\phi$ and $n_z = \cos\theta$. The free energy density for this deformation in the one constant approximation is given by

$$F = \frac{K}{2}[(\nabla\theta)^2 + (\sin\theta)^2(\phi_z^2 - 2q_0\phi_z + f)] \tag{18}$$

This leads to the following equations of equilibrium

$$\nabla^2\theta = \sin\theta\cos\theta[(\phi_z)^2 - 2q_0\phi_z + f] \tag{19}$$

$$(\sin\theta)^2\nabla^2\phi = -2\sin\theta\cos\theta[\nabla\theta \cdot (\phi_z - q_0)\hat{\mathbf{k}}] \tag{20}$$

These two coupled equations permit the following solutions in ϕ and θ

$$\phi = q_0 z \tag{21}$$

$$\theta - \frac{\pi}{2} = 2\tan^{-1}(\exp[\frac{z}{\eta_a}]) \tag{22}$$

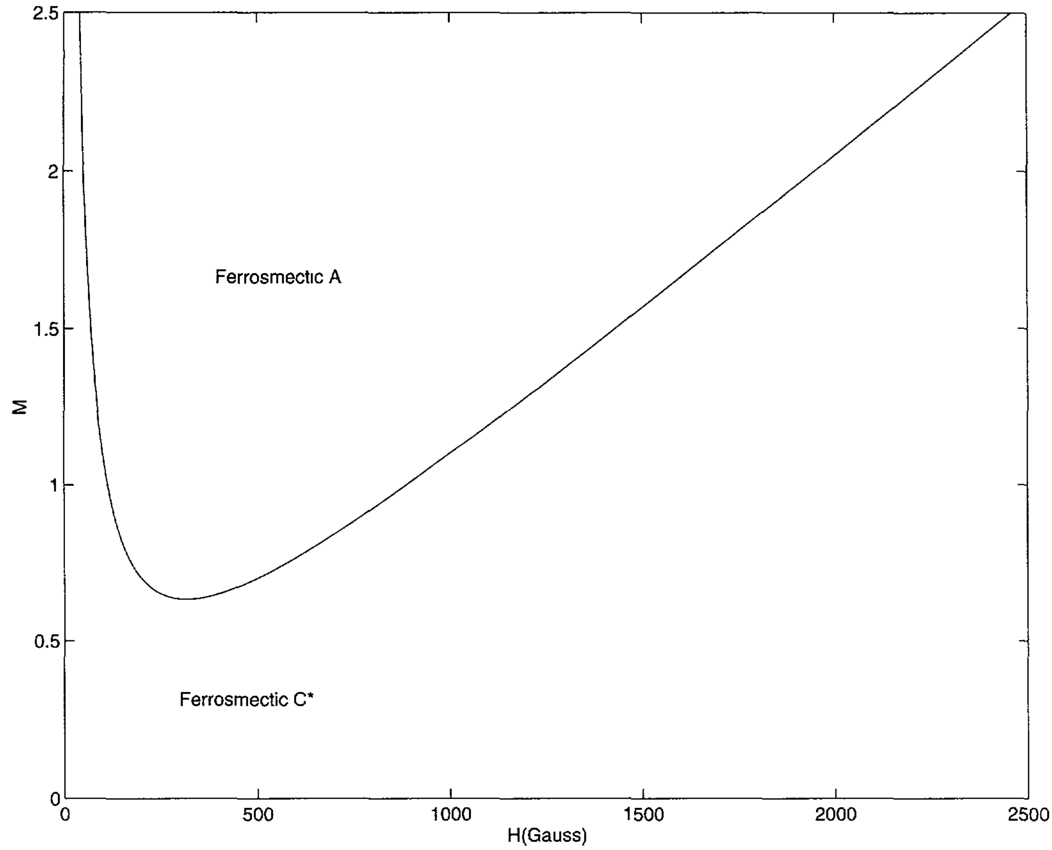

Fig. 4. — The phase diagram for the chiral FS_{C^*} phase. Here $\alpha_0 = 0.02$, $\chi_a = -0.1 \times 10^{-6}$, $\beta = 0.1$ and $\mathbf{M}_z$ and $\mathbf{H}$ parallel to one another. $\mathbf{M}$ is in units of 10^4 Gauss.

where $\eta_a = \sqrt{1/(q_0^2 - f)}$. Equation (22) describes a planar soliton of width $2\eta_a$ with $\theta(-\infty) = \pi/2$ and $\theta(+\infty) = 3\pi/2$. Within this width the director goes out of the cholesteric plane and at the centre of the soliton the director is along the twist axis. The structure of this soliton is depicted in Figure 5. We call this a 'Pinch Soliton' since the cholesteric lattice is pinched so to say in a narrow region of space. The width $2\eta_a$ of the pinch soliton, grows as the field increases and it diverges at a critical field given by $H_c = (2\pi/P)\sqrt{K/\chi_a}$. Hence we find a transition from the cholesteric state to the nematic state with the director $\mathbf{n}$ everywhere along the twist axis of the parent cholesteric. It is important to mention here that the energy required to create such a pinch soliton gradually decreases and goes to zero as H increases to H_c. Unlike the case of soliton lattice mediated transition [11, 12], the pitch in the present geometry does not change. We have a single soliton which grows in size and irons out the entire lattice at the critical field H_c. Interestingly this critical field is nearly 2/3 of that obtained in soliton lattice mediated transition.

It is important to note that in this pinch soliton shown in Figure 5, ϕ and θ variations are in the same direction namely the twist axis. Hence we call this a longitudinal pinch. Equation (19) also permits a soliton solution with θ varying in a direction perpendicular to the twist axis. Here ϕ continues to vary along z. This transverse pinch soliton is shown in Figure 6. As

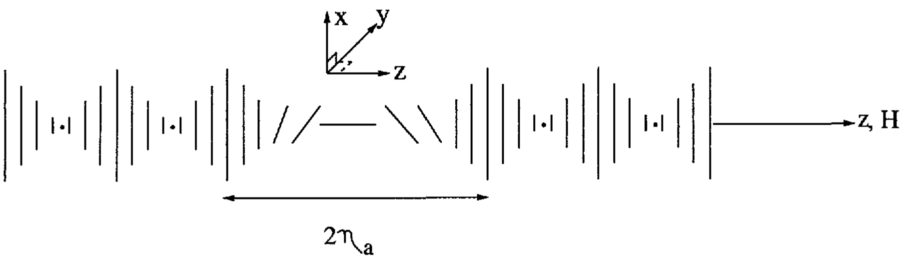

Fig. 5. — Structure of a longitudinal Pinch soliton. It has a θ variation along the twist axis.

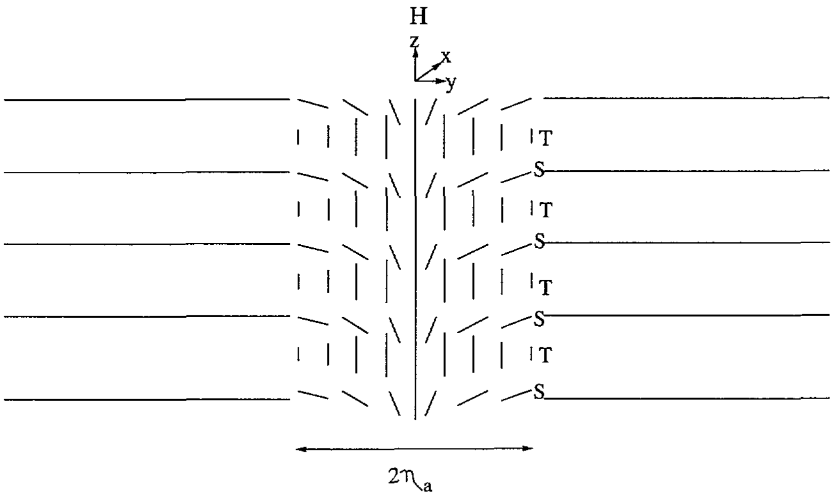

Fig. 6. — Structure of a transverse Pinch soliton with its θ variation perpendicular to the twist axis. T and S represent twist and splay-rich solitons respectively.

can be seen from the figure it is an alternate stack of twist and splay-rich solitons. The Ch-N change can be brought about through the creation of either a longitudinal or a transverse pinch soliton. In this simple model we cannot assert as to which mode of transformation the system will adopt. However it is not difficult to see that in the presence of elastic anisotropy one of the pinch solitons will be of higher energy compared to the other. Therefore in a real system there will be no ambiguity.

2.3.2. Chiralising Soliton in a Nematic. — We now consider the phase change that can be effected from the nematic side. From the discussion of the previous section we conclude that in fields parallel to the twist axis at $H > H_c$ a cholesteric becomes a uniform nematic with the director everywhere along the twist axis of the parent cholesteric i.e., $\mathbf{n_0} = (0,\ 0,\ 1)$. We can create a soliton in this nematic state also. With $\mathbf{n} = (\sin\theta\cos\phi,\ \sin\theta\sin\phi,\ \cos\theta)$ the free energy density is again given by (18) but with $H > H_c$ i.e., $f > q_0^2$. The equations of equilibrium then permit the following solutions

$$\phi = q_0 z \tag{23}$$

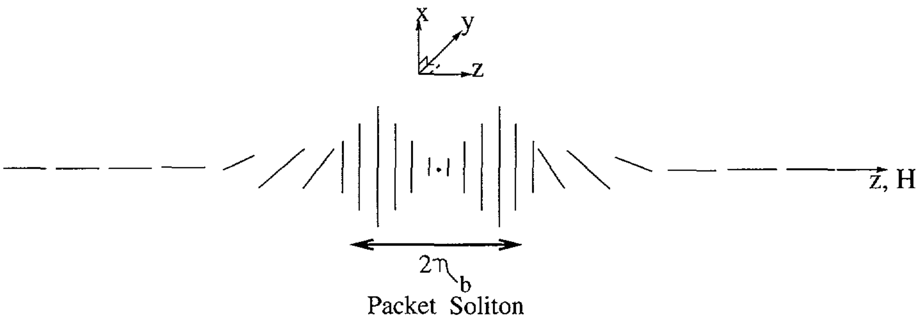

Fig. 7. — Structure of a longitudinal Packet soliton in a nematic.

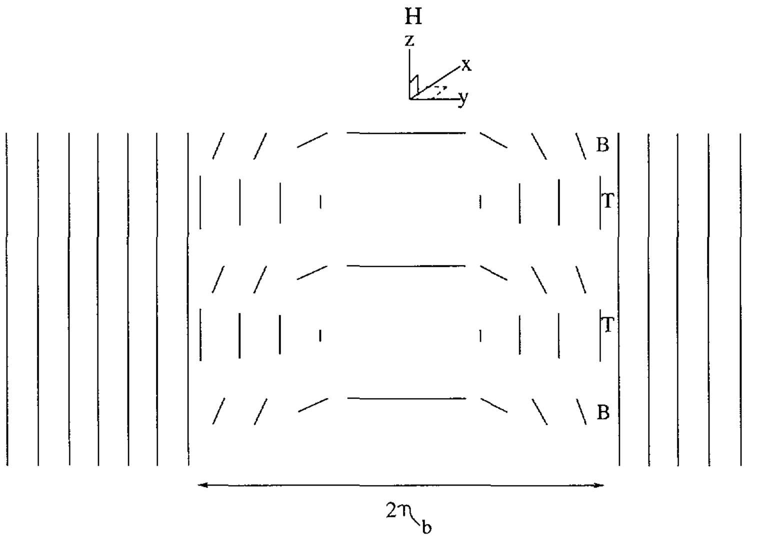

Fig. 8. — Structure of a transverse Packet Soliton in a nematic. T and B represent twist and bend-rich solitons respectively.

$$\theta = 2\tan^{-1}(\exp[\frac{z}{\eta_{\mathrm{b}}}]) \tag{24}$$

where $\eta_{\mathrm{b}} = \sqrt{1/(f - q_0^2)}$. Equation (24) describes a soliton which has a chirality as given by (23) with $\theta(-\infty) = 0$ and $\theta(+\infty) = \pi$. Its structure is schematically shown in Figure 7. Over a length of $2\eta_{\mathrm{b}}$ the uniform state can be distorted to form a cholesteric like section. We call this a 'Packet Soliton' since a lattice is packed inside this soliton. This lattice has a pitch of $2\pi/q_0$. On decreasing the field this region of width $2\eta_{\mathrm{b}}$ grows and at the critical field given by $H_{\mathrm{c}} = (2\pi/P)\sqrt{K/\chi_{\mathrm{a}}}$ the entire structure transforms into a cholesteric. In this case also, structural change is through a single soliton and the energy of its creation continuously goes to zero as H decreases to H_{c}.

As in the case of a pinch soliton, a packet soliton with θ variations in a direction perpendicular to the twist axis is also possible. This is schematically shown in Figure 8. This can be considered as an alternative stack of twist and bend-rich solitons. In the one constant approximation the widths and energies of both the transverse and longitudinal packet solitons are the same. Here also elastic anisotropy will decide as to which packet soliton triggers chirality in the nematic phase.

2.3.3. Ferrocholesteric to Ferronematic Transition. — In the case of the FCh-FN transition also we can construct a similar single soliton. The field free structure is described by $\mathbf{n_0} = (\cos\phi_0, \sin\phi_0, 0)$ with $\phi_0 = 2\pi z/P$. When the field is along z, a distortion described by $\mathbf{n} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ sets in. Here we get

$$\phi_z = q_0 \tag{25}$$

$$\frac{\partial^2\theta}{\partial z^2} = -(q_0^2 - f)\sin\theta\cos\theta + g\sin\theta \tag{26}$$

Clearly θ obeys a double sine-Gordon equation whose solutions are well known [4, 13]. For $f < q_0^2$ we get a soliton solution which is a combination of two solitons of winding numbers $2\theta_0$ and $2\pi - 2\theta_0$ respectively with $\theta_0 = \cos^{-1}[g/(q_0^2 - f)$. They are called respectively the N and W solitons [4, 13]. At $f = q_0^2$ these two solitons combine to give a 2π soliton and on further increase of the field this soliton will split at $(f - q_0^2) = g$ into two π solitons. But this cannot be ironed out by a continuous deformation at any higher finite field. Further at any field there will also be magnetic grain migration associated with the soliton structure. So a single soliton mediated FCh-FN transition is not a feasible alternative to the solution discussed in Section 2.1.

2.4. REMARKS. — It may be mentioned that all the structural transitions discussed in this section are permitted solutions to the equations of equilibrium. The solutions are such that a uniform twist exists in the medium even in the presence of an external field. However, there could be other solutions with a non-uniform twist and a different θ variation. These may even have lower energies. Hence the structural transition suggested by us, in any particular case should be looked upon as one of the possible modes of transition from a twisted configuration of the director field to the untwisted one and *vice-versa.*

3. Transitions in Crossed Electric and Magnetic Fields

So far we considered transformations in a magnetic field acting along the symmetry axis. The process of chiral-achiral transition will be very different in a magnetic field perpendicular to the symmetry axis. This has already been discussed in literature for cholesterics [11, 12], ferrocholesterics [3, 4] and for S_{C^*} [15–17]. In the case of S_{C^*}, the transitions have been considered in the neighbourhood of $S_A - S_{C^*}$ point. These transitions are mediated by the creation of soliton lattices which at a critical field go over to the achiral S_A or S_C phase. Here we consider the same phase transitions but in crossed electric ($\mathbf{E}$) and magnetic ($\mathbf{H}$) fields.

3.1. **E** AND **H** PERPENDICULAR TO THE TWIST AXIS. — We consider transitions in Ch and FCh systems in crossed electric and magnetic fields both in a plane perpendicular to the symmetry axis *viz.*, the twist axis.

3.1.1. Cholesterics. — Consider a cholesteric with a magnetic field $\mathbf{H}$ along the x axis and an electric field $\mathbf{E}$ perpendicular to it along the y axis. The director configuration is described by $\mathbf{n} = (\theta\cos\phi, \theta\sin\phi, 1)$. The free energy density is given by

$$F = \frac{K}{2}[(\phi_z^2 - 2\phi_z q_0)] - \frac{\chi_a}{2}H^2\cos^2\phi - \frac{\hat{\epsilon}_a}{2}E^2\sin^2\phi \tag{27}$$

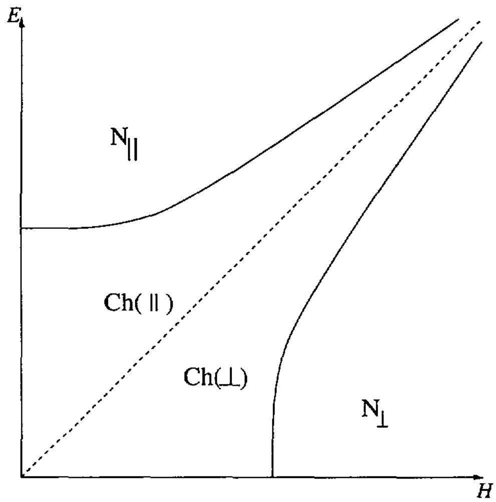

Fig. 9. — The schematic phase diagram for a cholesteric in crossed fields. $N_{||}$ and $N_\perp$ denote nematics which are aligned parallel and perpendicular to the H field respectively. Ch(||) denotes a cholesteric soliton lattice with nematic regions aligned parallel to the field and Ch(⊥) denotes that which has the nematic regions aligned perpendicular to the field.

where $\hat{\epsilon_a} = \epsilon_a/(4\pi)$ with ϵ_a as the dielectric anisotropy. Minimization of the total energy gives

$$\phi_{zz} = \frac{(\chi_a H^2 - \hat{\epsilon_a} E^2)}{K} \sin\phi\cos\phi \tag{28}$$

This is similar to the equation found in the usual de Gennes-Meyer transition in cholesterics. Hence the transition is driven by the formation of a π soliton lattice which on increase of either electric or magnetic field goes over to a nematic state aligned along the magnetic field ($N_{||}$) or to a nematic state aligned perpendicular to the magnetic field ($N_\perp$) depending upon whether $\chi_a H^2$ is more or less than $\hat{\epsilon_a} E^2$. The phase diagram is schematically shown in Figure 9. Here Ch(||) represents a soliton lattice where the nematic regions are parallel to the magnetic field and Ch(⊥) represents the one where the nematic regions are perpendicular to the magnetic field. As can be seen from Figure 9, change of the nematic phase from the $N_{||}$ to the $N_\perp$ state or *vice-versa* is possible. The cholesteric-nematic phase boundaries are given by

$$\pm\,(\chi_a H^2 - \hat{\epsilon_a} E^2) = \frac{K q_0^2 \pi^2}{4} \tag{29}$$

3.1.2. Ferrocholesterics. — We consider a ferrocholesterics (FCh) in the same geometry of crossed fields. Here we have to solve numerically two coupled differential equations one for ϕ distortions and another for grain migration. A very similar problem has already been considered [4]. We summarize here its implications since its generalization to the present problem is trivial.

We find that FCh to FN transition takes place as shown schematically in Figure 10 for $\chi_a > 0$ and $\epsilon_a > 0$. The FCh goes to the FN state either through the sequence of a 2π lattice followed by a split 2π i.e., $\pi - \pi$ lattice and ultimately to a $N_{||}$ ferronematic or through the sequence of a 2π lattice followed by an N – W lattice and finally to a $N_\perp$ ferronematic state. The transformation of the 2π lattice to either $\pi - \pi$ lattice or N – W lattice takes place along the phase boundaries

$$MH = \pm(\chi_a H^2 - \hat{\epsilon_a} E^2) \tag{30}$$

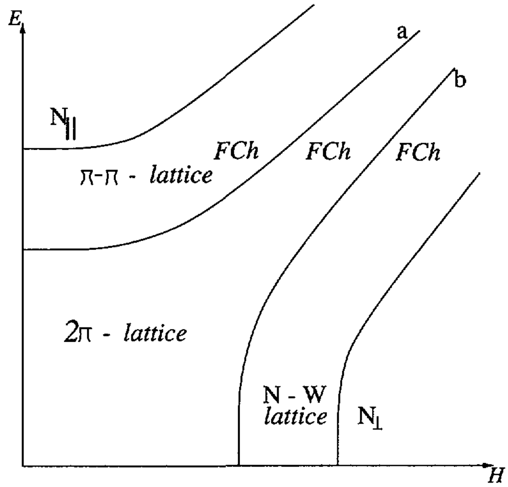

Fig. 10. — Schematic phase diagram for an FCh in crossed fields. Here a denotes the phase boundary $MH = \chi_a H^2 - \epsilon_a E^2$ and b denotes $MH = \epsilon_a E^2 - \chi_a H^2$.

These lattices on further increase of the field go over to $N_{||}$ or $N_\perp$ ferronematics. This phase boundary can only be numerically evaluated. It may be mentioned that for $\chi_a < 0$ and $\epsilon_a < 0$ ferrocholesterics, the regions of $\pi - \pi$ lattice and N − W lattice get interchanged in the phase diagram. Also the grain profiles for $\pi - \pi$ and N − W soliton lattices are entirely different [4]. Even here change of the nematic state from $N_{||}$ to $N_\perp$ and *vice-versa* is possible. In a similar way we can also discuss the case of χ_a and ϵ_a being of opposite signs.

3.2. **H** Along and **E** Perpendicular to the Symmetry Axis. — We now consider chiral-achiral transformations in FS_{C^*} with **H** along and **E** perpendicular to the symmetry axis i.e., **H** = (0, 0, H), **E** = (E, 0, 0) and **n** = $(\theta\cos\phi, \theta\sin\phi, 1)$. Here again we assume θ to be small. The free energy density without grain migration is

$$F = \frac{\alpha}{2}\theta^2 + \frac{1}{4}\beta\theta^4 + \frac{K}{2}[(\theta_z)^2 + \theta^2((\phi_z)^2 - 2q_0\phi_z)] + \frac{1}{2}\chi_a H^2\theta^2 - \frac{1}{8\pi}\epsilon_a E^2\theta^2(\cos\phi)^2 \pm \frac{MH}{2}\theta^2 \quad (31)$$

Phase transition due to a similar free energy density has been worked out by Michelson [14] and Yamashita [15–17]. We can easily extend their results to the present problem. We find that this system has phase diagrams which are interesting variations of those obtained by Michelson [14] and Yamashita [15–17]. Two of the very interesting possible phase diagrams are shown schematically in Figures 11 and 12. These are respectively for $\epsilon_a > 0$ and $\epsilon_a < 0$ materials. We find that this system can exhibit the features of reentrance together with tricritical and Lifshitz points. The essential features of this phase diagram can be easily understood. At $(\alpha + \chi_a H^2 + MH) << 0$ we can expect what Yamashita and Michelson predict in the low temperature region of S_{C^*} i.e., a second order FS_{C^*} to FS_C transition. In the neighbourhood of $(\alpha + \chi_a H^2 + MH) = 0$, this transition becomes first order resulting in a tricritical point C on the $FS_{C^*} - FS_C$ phase boundary. At $(\alpha + \chi_a H^2 + MH) >> 0$ this phase boundary meets the $FS_C - FS_A$ phase line tangentially at the Lifshitz point L. These arguments hold good for both $\epsilon_a > 0$ and $\epsilon_a < 0$ case as well.

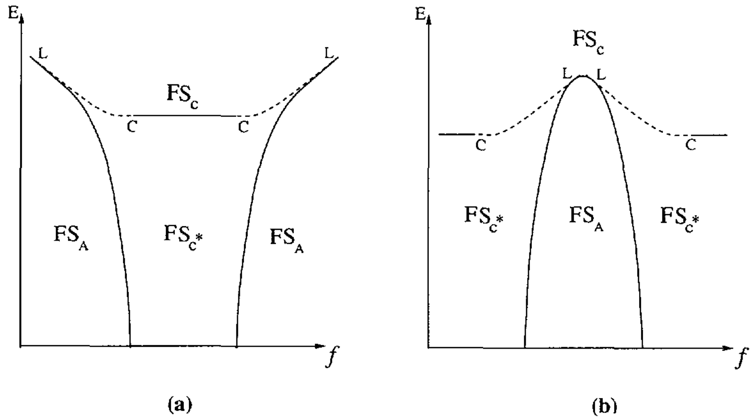

Fig. 11. — A possible schematic phase diagram of a $\epsilon_a > 0$ ferrosmectic in crossed fields. For (a) $\alpha > 0$, $\chi_a > 0$ and $\mathbf{M}{\cdot}\mathbf{H} < 0$. (b) $\alpha < 0$, $\chi_a < 0$ and $\mathbf{M}{\cdot}\mathbf{H} > 0$. The full line represents second order phase transition and the dashed line the first order transition. Points C and L represent tricritical and Lifshitz points. Here $f = (\chi_a H^2 + MH)$.

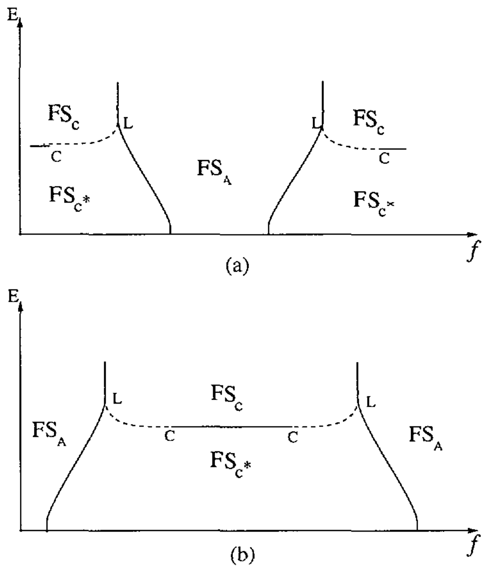

Fig. 12. — A possible schematic phase diagram for $\epsilon_a < 0$ ferrosmectic in crossed field. The notations are the same as those of Figure 11. For (a) $\mathbf{M}{\cdot}\mathbf{H} > 0$, $\alpha < 0$, $\chi_a < 0$ and for (b) $\mathbf{M}{\cdot}\mathbf{H} < 0$, $\alpha > 0$ and $\chi_a > 0$. Here $f = (\chi_a H^2 + MH)$.

3.3. REMARKS. — We have intentionally not considered the following geometries in our study.

i) FS_{C^*} and FCh with **E** along the twist axis and **H** perpendicular to it.

ii) FS_{C^*} with **E** and **H** perpendicular to the twist axis.

iii) Cholesterics with **E** (or **H**) parallel to the twist axis and **H** (or **E**) perpendicular to the twist axis.

We make the following comments regarding these geometries. In geometry (i) due to the $\mathbf{M} \cdot \mathbf{H}$ term in the free energy density the FS_A phase will not exist. Also if $\chi_a = 0$, we can expect a phase diagram similar to that obtained by Yamashita [16] for a ferroelectric S_{C^*} with $\epsilon_a = 0$ and in an electric field parallel to the smectic planes. There will be no FS_A phase and the FS_{C^*} to FS_C phase boundary will have two tricritical points. However when $\chi_a \neq 0$ we can extrapolate the results of magnetic field effects on an FCh and electric field effects on a ferroelectric S_{C^*}. It has been shown that in the case of FCh, in a magnetic field perpendicular to the twist axis, to start with we get a 2π soliton lattice which transforms to either a $\pi - \pi$ soliton lattice ($\chi_a > 0$) or a $\mathrm{N} - \mathrm{W}$ soliton lattice ($\chi_a < 0$) at a certain field H. A very similar result can be expected in the case of FS_{C^*} also. This phase transition is second order. Therefore we expect a new phase boundary in the FS_{C^*} region before it goes over to FS_C. At low electric fields (acting along the twist axis) we can expect this transition to be still second order. However at high electric fields it will be different in view of the fact that the soliton structure is quite different in this region. Here even a single 2π soliton has ripples in its θ profile. Extending the arguments of Yamashita [17], we speculate that this leads to an attraction between like 2π solitons resulting in a first order transition from the 2π soliton lattice to a $\pi - \pi$ or $\mathrm{N} - \mathrm{W}$ soliton lattice. Therefore we expect on this new phase boundary a tricritical point as well. The way this new phase boundary meets the FS_{C^*}-FS_C boundary is not easy to speculate upon. All these features are plausible even in the case of an FCh in a similar geometry. In geometry (ii) the phase transition is qualitatively similar to an FCh in the same geometry. We conjecture that in geometry (iii), we can expect a phase diagram similar to that obtained by Yamashita [15] for ferroelectric S_{C^*} in a magnetic field along the layers. Here we will be having a phase transition from Ch to $N_{||}$ and Ch to $N_{\perp}$ states in the place of FS_C and FS_A states.

It should be emphasized that phase diagrams in all these cases can be constructed only by undertaking detailed and elaborate calculations pertaining to the structure and energetics of the soliton lattices. In this paper we have not addressed ourselves to this exercise.

4. Effect of Boundaries

It has been implicitly assumed in the case of FCh-FN and Ch-N transitions, that a global reorientation of the helical axis perpendicular to the field is prevented by sample boundaries. In the case of FS with $\mathbf{M}$ anti-parallel to $\mathbf{H}$, a global flip of the sample to the configuration of $\mathbf{M}$ parallel to $\mathbf{H}$ is again assumed to be prevented by the sample boundaries. In this context a few remarks on the boundary effects are in order ([1]).

In Ch and FCh systems we can easily realise in the laboratory two boundary conditions viz., the twist axis is either parallel or perpendicular to the bounding surface. In FS systems likewise, we have two boundary conditions viz., smectic layers are either parallel or perpendicular to the bounding surface. In such situations our values of θ and ϕ should be matched smoothly with the values of θ and ϕ existing at the boundaries of the sample. This takes place over a coherence length in the neighbourhood of the sample boundaries. The value of the coherence length depends upon the field and elastic constants. Though this can be explicitly included in each problem we may still expect many of our solutions to be reasonably valid in large enough samples under appropriate boundary conditions. In particular, we make the following observations:

(1) In the case of FS we can easily orient the layers but cannot anchor θ or ϕ at the boundaries. Hence for both the boundary conditions solutions discussed under Section 2.2 will be valid.

([1]) We are thankful to the referees for comments

(2) The *longitudinal pinch* soliton solution suggested in the case of Ch to N transition dealt with in Section 2.3.1, is a natural solution that is compatible with the boundaries at which the twist axis is normal to the walls. Similarly the *transverse pinch* soliton can be expected as a natural solution in the case of samples where the boundaries orient the twist axis parallel to the walls.

(3) In all the situations in Section 3, where the field induces a soliton lattice the appropriate boundary condition to be chosen is that where the twist axis is perpendicular to the wall or where the smectic layers are parallel to the walls. Then all the solutions discussed under this section are valid.

(4) In all the other cases, the solutions obtained can be matched with either of the boundary conditions viz., the twist axis is parallel or perpendicular to the walls. This matching can be effected over a coherence length near the bounding surfaces. Further, for FN to FCh transition treated in Section 2.1.2, for both boundary conditions, the director is already predisposed to tilt in a particular direction. Hence this transition will not be defect mediated.

5. Conclusion

We have studied field induced chiral-achiral phase transitions in some liquid crystals. In the defect mediated transitions we have considered transformations triggered by disclinations, single solitons and soliton lattices. We have also discussed transitions not involving defects. Many interesting results have been obtained with the phenomenon of reentrance, tricritical points and Lifshitz points accompanying these transitions.

Acknowledgments

Our thanks are due to K.A Suresh for useful comments.

References

[1] Kutnjak-Urbane. B. and Zeks B., "Phase Transitions in Liquid Crystals", *NATO Adv. Study Inst. Series*, S. Martelluci and A.N. Chester Eds. (Plenum, N.Y. 1992).
[2] Chen S.H. and Amer N.M., *Phys. Rev. Lett.* **51** (1983) 2298.
[3] Brochard F. and de Gennes P.G., *J. Phys. France* **31** (1970) 691.
[4] SunilKumar P.B. and Ranganath G.S. , *J. Phys. II France* **3** (1993) 1497.
[5] SunilKumar P.B. and Ranganath G.S.,*Mol. Cryst. Liq. Cryst.* **196** (1991) 27.
[6] Friedal G. *Ann. Phys* **18** (1922) 273; See also Ranganath G.S., Suresh K.A., Rajagopalan S.R and Kini U.D *Pramana, Suppl.* (1975) 353.
[7] Dierking I., Giebelmann F., Zugenmaier P., Mohr K., Zaschke H. and Kuczynski W., *Liquid Cryst.* **18** (1995) 443.
[8] Ponsinet V., Fabre P., Veyssie M. and Cabanel R., *J. Phys. II France* **4** (1994) 1785.
[9] Ponsinet V., Fabre P. and Veyssie M. , *Europhys. Lett.* **30** (1995) 277.

[10] Ranganath G.S. , *Mol. Cryst. Liq. Cryst. Lett.* **92** (1983) 201.
[11] de Gennes P.G . , *Solid State Commun.* **6** (1968) 163.
[12] Meyer R.B., *App. Phys. Lett.* **12** (1968) 281.
[13] Dimitrienko V.E. and Belayakov V.A., *Sov. Phys. JETP* **51** (1980) 787.
[14] Michelson A., *Phys. Rev. Lett* **39** (1977) 464.
[15] Yamashita M., *J. Phys. Soc. Jpn* **57** (1988) 2051.
[16] Yamashita M., *J. Phys. Soc. Jpn* **56** (1987) 1414.
[17] Yamashita M., *Prog. Theor. Phys* **74** (1985) 622.