Nonlinear Optical Effects
in
Liquid Crystals

by

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As when the mantra sinks in Yoga's ear,
Its message enters stirring the blind brain
And keeps in the dim ignorant cells its sound;
The hearer understands a form of words
And, musing on the index thought it holds,
He strives to read it with the labouring mind,
But finds bright hints, not the embodied truth:
Then, falling silent in himself to know
He meets the deeper listening of his soul:
The Word repeats itself in rythmic strains:
Thought, vision, feeling, sense, the body's self
Are seized unalterably and he endures
An ecstasy and an immortal change;
He feels a Wideness and becomes a Power,
All knowledge rushes on him like a sea:
Transmuted by the white spiritual ray
He walks naked heavens of joy and calm,
Sees the God-face and hears transcendent speech:
An equal greatness in her life was sown.

Sri Aurobindo in Savitri, -p375

CERTIFICATE

This is to certify that the thesis entitled **Nonlinear Optical** Effects **in Liquid Crystals** submitted by *Srivatsa S* K for the award of the degree of DOCTOR OF PHILOSOPHY of Jawaharlal Nehru University is his original **work.** This has nor been published or submitted to any other university for any other degree or diploma.

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DECLARATION

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

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The library at RRI must have a special mention in this acknowledgement. The book collections and its maintenance is really unique. Every visit to the library would transport one into a whole new world. The ever smiling staff of the library would oblige all our requests including books and journals from various other libraries in Bangalore and finally even xeroxing this thesis in a very short time. I sincerely thank all the staff of our library.

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John Archbald Wheeler, of the MTW fame for Relativists, said of his students: 'students are there to teach the professors'! I would say similarly with respect to my juniors. I came across wonderful friends, as my juniors, who have taught me many of their different specialities in physics. Discussions with Sushil, Rekesh, Panikumar, Vishwanath and others on various topics kept the environment lively. Also I had the opprtunity to learn the 'tricks of the trade' from my seniors. The much desired help in numerical work was provided by Sreejith and Dipanjan.

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The works of Sri Aurobindo and The Mother have been a constant inspiration in my life. These studies immensely enriched the quality of my life in general and my intellectual life in particular. I dedicate this work in all humility to *Sri Aurobindo* and *The Mother*.

Contents

LIST OF SYMBOLS

Every symbol was a reality
And brought the Presence which have given it life..
Sri Aurobindo in Savitri

$\mathbf{n}$	Nematic director
N	Layer Normal in smectics
S	Nematic order parameter
ϕ	Angle between the director and the external field
θ	Tilt of the molecule with respect to the layer normal in smectic C
$P_\circ$	Pitch of the cholesteric
P	Pitch of the twisted structure
$\mathbf{q}_\circ$	Wave vector of periodicity
q_i	Wave vector components
$\epsilon^s_\perp$	Static dielectric constant perpendicular to the director
$\epsilon^s_\parallel$	Static dielectric constant parallel to the director
$\epsilon^s_a = \epsilon^s_\parallel - \epsilon^s_\perp$	Static dielectric anisotropy
$\mathbf{P_{flexo}}$	Flexoelectric polarisation
$e_1,\ e_3, e$	Flexoelectric constants
E	Static electric field
$\chi_\parallel$	Diamagnetic susceptibility component parallel to the director
$\chi_\perp$	Diamagnetic susceptibility component perpendicular to the director
$\chi_a = \chi_\parallel - \chi_\perp$	Diamagnetic anisotropy
H , H	Magnetic field vector and its strength
ξ	Coherence length
$\eta = z/\xi$	
α_1, α_2	Phenomenological constants in the free energy expansion of smectics
m	Magnetisation of an individual magnetic grain
Ω	Volume of the sample
$\rho = m\Omega/(k_B T)$	
f	Magnetic grain concentration
a	Scattering cross-section

ϵ_{ij}	Optical dielectric tensor components
$\epsilon_{\|\|}$	Optical dielectric constant parallel to the director
$\epsilon_{\perp}$	Optical dielectric constant perpendicular to the director
$\epsilon_a = (\epsilon_{\|\|} - \epsilon_{\perp})$	Optical dielectric anisotropy
μ	Refractive index
$\mu_{\|\|}$	Refractive index parallel to the director
$\mu_{\perp}$	Refractive index perpendicular to the director
$\Delta\mu$	Local birefringence
c	Velocity of light
$\mathcal{E}$	Electric vector of the laser beam
I	Laser intensity
I_{th}	Threshold laser intensity
w	Angular frequency of light
λ	Wavelength of light
$k = \frac{2\pi}{\lambda}\mu$	, Wave vector of light in the medium
K_1	Splay elastic constant
K_2	Twist elastic constant
K_3	Bend elastic constant
K	Curvature elastic constant in the one constant approximation
u	Smectic layer dispacement
B	Elastic constant corresponding to smectic layer compression or dilation
K	Effective curvature elastic constant of a cholesteric in the coarse-grained description
$\mathcal{F}_{elastic}$	Elastic free energy density due to director distortions
$\mathcal{F}_{optical}$	Optical free energy density
$\mathfrak{Z}$	Free energy density
F_{mag}	Diamagnetic free energy density in a nematic
F_{elec}	Dielectric free energy density in a nematic
$\mathfrak{Z}$	Coarse-grained free-energy density in cholesterics
$\delta\mu$	Induced change in refractive index
Φ	Phase of a dark soliton
μ_{nl}	Nonlinear change in refractive index as a function of intensity

η_1	Nonlinear coefficients due to the laser supression of director fluctuations
η_2	Nonlinear coefficients due to the laser induced tilt in smectics
ψ	Real amplitude of the fields in an optical soliton
γ	Ratio of nonlinear coefficients η_1 and η_2
ν	Real parameter related to periodicity of soliton along the longitudinal direction
κ	Average thermal conductivity of a liquid crystal
χ	Optical absorption coefficient
l	Molecular size
x, y, z	Coordinate axes
$\mathbf{r}$	Coordinate vector
X, Z	Scaled coordinates
G	Gauss
k_B	Boltzmann constant
T	Absolute temperature
t	Sample thickness
Q	Tricritical point

PREFACE

Liquid Crystals are "soft" materials. Their response to even weak mechanical stresses, electric or magnetic fields is very large. They possess many physical properties typical of both liquids and crystals. For instance, they flow like liquids and also exhibit optical and other anisotropic properties generally associated with crystals. A laser beam also influences the structure and properties of the liquid crystals through its electric field. For instance, it leads to very large changes in the refractive index of the medium resulting in very pronounced nonlinear optical effects. Nonlinear optics, in general, is the study of physical processes leading to laser induced changes in the medium and the reciprocal influence of these changes on the laser beam itself. This thesis deals with such nonlinear optical studies in liquid crystals.

Liquid Crystals possess not only a structural symmetry that is in between that of crystals and the usual liquids but also occur as thermodynamic phases between these familiar states of matter. Hence, they are often referred to as mesophases. These mesophases are observed only when the constituent molecules have a large shape anisotropy, i.e., these molecules are either rod-like or disc-like. Further, liquid crystals exhibit anisotropy in their physical properties. One of the extensively studied liquid crystalline phase is the *nematic* which has a preferential alignment of its rod-like or disc-like molecules. The average direction of the orientation in case of rod-like molecules is described by a unit vector called the director. In the discotic nematic the disc-like molecules are aligned nearly perpendicular to the director. The nematic phase possesses only long range orientational molecular order and has no long range positional molecular order. If in addition the constituent molecules are chiral or if chiral dopants are added to a nematic then there will be a macroscopic twist of the director in a direction perpendicular to the director. These chiral liquid crystals are called *cholesterics*. Generally the pitch of the cholesteric structure will be between 0.1 to 100 μm. We also find positional order in some liquid crystals. For example, in a liquid crystal with layered arrangement we have a one dimensional positional ordering along the director. These are called *smectic* liquid crystals. If the director is normal to the layers it is called smectic *A.* If the director is ar an angle to the layer we get a *smectic* C. We briefly review in the first chapter of this thesis the Structure, Optics, Elasticity and Nonlinear Optics of liquid crystals. This would provide the background material for the new results reported in the succeeding chapters.

The interest in the nonlinear optics of liquid crystals is mainly due to the observation that their nonlinear optical coefficients are invariably very large. Liquid crystals being optically anisotropic, the electric field of an intense laser beam leads to a dielectric torque which can reorient the director. This results in a change of the refractive index as seen by the laser beam. In fact. it is found that the nonlinear coefficient for a change of the refractive index is six orders of magnitude higher than what we find even in the so called highly nonlinear materials, like for example CS_2. This is the reason why liquid crystals are said to possess giant optical nonlinearities. Hence, nonlinear optical effects can be observed in liquid crystals even at low laser intensity levels. Thus liquid crystals become the most appropriate materials for testing the various general predictions of nonlinear optics that are difficult to demonstrate or study in usual nonlinear materials. Director reorientation is not the only process operating in such liquid crystals. We find that there are two other new nonlinear optical processes that can exist in liquid crystals. viz., the *laser suppression of the director fluctuations* and *laser induced tilt angle in smectics*. We also study the possible new nonlinear optical effects due to these processes.

It was pointed out in the 70's by P. G. de Gennes that an external static magnetic field acting along the director of a nematic can suppress its thermal fluctuations thus enhancing the dielectric anisotropy of the medium and in particular to an increase(decrease) in the dielectric constant parallel(perpendicular) to the direcror. In chapter 2 we generalise this idea by considering the suppression of the director fluctuations in a nematic in the electric field of a laser beam. Some of the consequences of this mechanism are:

- The increase in the dielectric tensor component along the director is proportional to the square root of the laser intensity. This result should be contrasted with the familiar nonlinear processes where a change in refractive index is proportional to the intensity of the laser beam. The effective change in the refractive index is of the order of 10^{-4} for an intensity of 10 kW/cm^2. On the other hand, at the same intensity the usual classical Kerr nonlinearity leads to a change in refractive index of only 10^{-6}. Thus the optical nonlinearity due to this new process is considerably larger.

- A partially polarised light beam of finite width with its intense coherent component parallel to the director, having a central peak intensity profile, effectively suppresses the director fluctuations more at the center than at its edges. Hence, this component is self-focused. On the other hand, the orthogonal weak incoherent component does not affect the director fluctuations in the medium but is affected by the completely polarised component which actually diminishes the refractive index for this component. The effect being more at the center. Therefore this orthogonal component undergoes self-divergence.

- In a confined nematic with the director parallel to the boundaries, we consider light propagating with its electric vector parallel to the director. The wave reflected from the rear boundary and the incident wave interfere to set up a standing wave in the medium. Gradually the intensities at the antinodes increase. Finally we get a periodic variation in the refractive index. Interestingly, its periodicity satisfies the Bragg condition for the reflection of the incident wave. This leads to self-induced Bragg reflection or self-iridescence.

- Generalisation of the process of laser suppression of the director fluctuations to cholesteric liquid crystals leads to some new optical effects. When the wavelength of light is very small compared to the pitch of the cholesteric a permitted eigenmode of the laser beam has its electric vector strictly either parallel or perpendicular to the director everywhere. It is experimentally possible to select the mode with the electric vector along the director and the said process will become important. Thus we get an increase in anisotropy of the local refractive index or in effect the order parameter. This leads to an increase in the twist elastic constant. Hence the structure unwinds to some extent. Our calculations indicate that this is large enough to be experimentally detectable.

- Now we come to the Bragg reflection mode of cholesterics. Complete reflection of an incident circularly polarised light takes place over a band of wavelengths. In a right(left) handed helix the right(left) circularly polarised beam is totally reflected and the left(right) circularly polarised light is transmitted. Inside the medium the totally reflected wave and the incident wave interfere to form a standing wave which is locally linearly polarised. In right handed cholesterics at the long wavelength edge of the optical Bragg band the electric vector of this linearly polarised standing wave is parallel to the local director. Thus we can invoke at this wavelength the process of laser suppression of the director fluctuations. In view of what has been said earlier this leads to local unwinding. Since the electric field of the standing wave decays inside the medium, structural changes due to this process also decreases from the surface. Therefore, complete phase-matching between the incident and the reflected waves is affected. This reduces the amplitude of the linearly polarised standing wave. The local optical effect on the structure decreases. Thus the system relaxes towards its zero field structure leading to a better phase-matching between the incident and the reflected waves. However, this in turn increases the amplitude of the standing wave. This process repeats itself. Therefore, the system exhibits temporal oscillations in the twist of the structure (near the surface) and the transmitted intensity.

In chapter **3** we briefly dwell upon another new optical nonlinearity. Laser fields can also change the tilt of the molecules relative to the layer normal in smectic liquid crystals. The resultant change in the tilt angle changes the refractive index as seen by the laser beam. Like the normal Kerr process the change in the refractive index is linearly dependent on the laser intensity. But its magnitude is extremely large, and comparable to the giant nonlinearity associated with the director reorientation. Further, this nonlinear coefficient is positive. Thus a new nonlinear optical process can operate in smectic liquid crystals. Some of the interesting consequences of this process are:

- In the smectic A phase a laser beam propagating along the layer normal with its electric vector parallel to the layers induces a smectic A to smectic C transition, beyond a threshold intensity. This threshold intensity depends on how far the system is away from the same transition that can be induced thermally. The intensity required for this process is quite low- of the order of $5\ kW/cm^2$.
- A chiral smectic C is a smectic C made of chiral molecules or with chiral dopants with a spontaneous twist along the layer normal. Generally, the pitch and the tilt are coupled with the pitch being a non-monotonic function of the temperature. For an increase of temperature near the chiral smectic C to smectic A transition. the pitch initially increases, reaches a maximum and then decreases to zero at the transition point. Thus at any given temperature the incident laser induces an increase or decrease of the twist of the structure.
- In a standing wave set up in a confined smectic A, due to the interference between the incident and the reflected waves propagating along the layer normal, a tilt is induced at the antinodes. The periodic intensity distribution results in a periodic structure which has alternating smectic A and smectic C blocks. A similar structure is to be expected in a smectic A which has a low temperature chiral smectic C. Then we get twisted regions between smectic A blocks. Such structures are rather reminiscent of what are called the twist grain boundary smectics which have smectic like blocks separated by thin twist boundaries. On the other hand in a chiral smectic C phase near the transition to smectic A, a laser beam propagating parallel to the layers with its electric vector parallel to the helical axis results in a two dimensional periodic structure in a standing wave set up inside the medium.
- In a smectic A phase, the familiar Kerr nonlinear process can lead to the generation of a third harmonic. If the phase-matching condition is not achieved then the energy alternates between the fundamental and the third harmonic. At higher intensities the tilt induced is higher at the harmonic due to higher optical anisotropy at this wavelength. We again get a periodic structure.

From the early days of nonlinear optics of liquid crystals, the director reorientation in a laser beam was the only mechanism considered in this field. This was largely due to the associated high optical nonlinearities. This process remains by far the most studied and discussed by various groups throughout the world. We present in this thesis some new nonlinear optical effects due to this ven- process in both achiral and chiral liquid crystals. It is well known that a static electric or magnetic field also affects the director orientation through the dielectric or diamagnetic torques acting on the director. In chapter **4** we discuss the nonlinear optical effects in a nematic and a nematic doped with ferromagnetic grains. Incidentally: a dilute suspension of the ferromagnetic grains results in a ferromagnetic behaviour of a nematic and is referred to in the literature as a ferronematic. We consider the simultaneous presence of both a static electric or magnetic field and the optical field. The new and interesting results obtained here are:

- In a confined nematic with the director anchored at an angle to the boundaries, a linearly polarised laser beam incident normal to the glass-places polarised parallel to the plane of director tilt induces director reorientation. The standing wave set up inside the medium due to reflections from the rear boundary leads to spatially changing periodic director orientation. This periodicity naturally satisfies the Bragg condition and results in Bragg reflection or self-iridescence as it happened in the case of laser suppression of the director fluctuations in a uniform nematic.
- In the presence of a static magnetic field and the optical field of a laser acting orthogonal to each other in a nematic, the phase diagram of transitions between the allowed states of the uniform director orientation has been worked out. The transition from a uniform state parallel to the magnetic field to one parallel to the electric vector of the laser is of first order. Hence these two uniform states can coexist over a range of magnetic field strengths and laser intensities.
- The phase diagram for allowed base states in a ferronematic with the simultaneous action of both the magnetic field and an optical field have also been worked out. In the same geometry i.e., with the magnetic field perpendicular to the electric vector of the laser beam in a nematic but with a negative diamagnetic anisotropy, we

find that the system exhibits a reentrant phenomenon below a certain laser intensity.

- In a cholesteric, in the short wavelength limit, for two counter-propagating laser beams propagating parallel to its twist axis and polarised respectively parallel and perpendicular to the director, an increase in the intensity of one of the beams whose electric vector is perpendicular to the director, leads to a global structural rotation through 90°. This is in a sense a switching of orientation.

The study of propagation of laser fields through 1D lattices is the subject matter of chapter 5. We first consider light propagation in a nematic in the presence of a static electric field. Next we study the cholesteric in various geometries. The new results obtained here are:

- Generally, a static electric field in a nematic can induce planar periodic distortions. This is referred to as a flexoelectric (which is akin to the piezoelectricity of crystals) lattice in the literature. The formation of such a lattice crucially depends on the dielectric anisotropy and flexoelectric constants of the medium. In a nematic, in a static electric field with the material constants such that the lattice is not induced, we find that a suitably polarised laser beam can induce an instability leading to the formation of a flexoelectric lattice.
- A flexoelectric lattice naturally exhibits Bragg reflections for light propagating along the lattice vector with its electric vector along the static electric field. At the short wavelength edge of the Bragg band the electric vector at the antinodes of the standing wave is perpendicular to the director. The resulting director reorientation causes a phase mismatch between the forward and backward propagating waves. The dielectric torque decreases eventually resulting in a better phase matching. This process repeats resulting in self-induced oscillations of the structure and the transmitted intensity.
- A cholesteric to nematic transition can be induced by an external static electric or magnetic field acting perpendicular to the twist axis. With the increase in the field strength the pitch increases and at a critical field the helix unwinds completely. A similar effect is also possible in a laser beam propagating along the twist axis. If the

wavelength of the incident light is very much greater than the pitch of the helix then unwinding of the structure takes place initially. But eventually the pitch matches the wavelength, resulting in Bragg reflection, thus inhibiting a global unwinding of the helix.

When the beam propagates perpendicular to the twist axis of a cholesteric some of the important results obtained are:

- With the electric vector also perpendicular to the helical axis we get local director reorientation at places where the director is not parallel to the electric field. The induced structural distortions leads to a corrugation of an incident plane wavefront resulting in optical diffraction. Only in thin samples, where internal diffraction effects can be neglected, the cholesteric liquid crystals can be unwound at a critical intensity. In thick samples, however, complete unwinding is not possible.
- In the same geometry in the presence of Kerr nonlinearity we find that the dielectric tensor component parallel to the helix axis becomes a periodic function of space. This results in a new diffraction mode. A low intensity light beam polarised parallel to the helix axis and propagating perpendicular to the twist axis which would otherwise be not diffracted will now be diffracted as in a periodic phase grating.
- In a finite sample bounded and anchored at the walls when the electric vector of the laser is parallel to the helix axis an undulation instability sets in beyond a threshold intensity. This is in the nature of periodic structural distortions in the director perpendicular to the twist axis. For normal incidence the instability occurs at a low intensity of a few kW/cm^2 for samples of thicknesses $\approx$ 100 μm. A similar instability is also possible in the smectic $\boldsymbol{A}$ phase when a laser beam propagates perpendicular to the layers with its electric vector parallel to the layers. This instability leads to a periodic structural distortion and hence to a periodic refractive index variation across the wavefront of the incident beam resulting in a novel self-induced grating diffraction.

We discuss in chapter 6 structural solitons, also known as walls or kinks, in nematics. These arise because of a degeneracy in the director orientation in an external electric or magnetic field. As a result the director can be parallel or antiparallel to the static external field. This is due to the symmetry of a nematic director. The director turns through 180° along a direction normal to the wall. We study the nonlinear optical effects on such a wall. We consider the combined effect of both the static magnetic field and a laser field in nematics. The main results are :

- The phase transition between two types of allowed kink states is first order. The phase diagram here is identical to the uniform state phase diagram described earlier.
- There exists a new kink state with a topological winding of the director equal to $\pi/2$. Incidentally, such a kink is forbidden in static fields.
- In a ferronematic with negative diamagnetic anisotropy and with the magnetic field perpendicular to the electric vector of the laser beam, the transitions between two allowed kink states exhibits reentrant behaviour and a tricritical point. However, in the same geometry with positive diamagnetic anisotropy the phase diagram has a tricritical point. Reentrant behaviour is also predicted but over a narrow range of intensities.

In the last chapter (chapter 7), we consider the effects on the laser beam due to the various nonlinear processes operating in liquid crystals. We also consider change in refractive index due to heating of the sample by laser absorption. This is called Thermal Indexing of the medium. We consider two regimes: when the beam width is so large that self-diffraction can be neglected, and when the self-diffraction of the beam cannot be ignored. In the latter case we get optical spatial solitons. In case of very wide beams some of the important conclusions are:

- In the presence of nonlinearities due to suppression of the director fluctuations and thermal indexing, we find that the beam width is periodically modulated in space as it propagates through a nematic with its electric vector parallel to the director.
- An unpolarised beam with a central peak intensity profile, propagating perpendicular to the director in a nematic gets resolved into two components, one parallel to the director and another perpendicular to the director. Due to thermal indexing the component parallel to the director experiences a negative optical nonlinearity

and becomes a divergent beam. The component perpendicular to the director sees a positive optical nonlinearity and becomes a convergent beam. This, in thin samples, gives rise to a bright central spot with polarisation perpendicular to the director surrounded by a broad region polarised orthogonal to the central spot.

- In a confined nematic with the director perpendicular to the wall at one boundary and parallel to the wall at the other boundary(also called a hybrid geometry), due to thermal indexing alone, the beam width is affected. The material parameters and the thickness of the sample could be so chosen that the beam width returns to the original value on exit at the rear boundary. The same process in an unbounded flexoelectric lattice implies a periodic modulation of the beam width as the laser beam propagates along the direction of periodicity.

We lastly consider the spatial optical solitons where the nonlinearity due to various processes briefly described above act in conjunction but against the inevitable self-diffraction of the beam. Here, though they are referred to as solitons we must mention that all these are only solitary waves which do not share all the properties of the usual solitons, like preservation of shape in a pairwise collision. We work in the thin-film approximation where intensity reduction due to absorption is negligible over the sample thickness. Some of the important results of this investigation are:

- The structure of the spatial soliton due to the laser suppression of the director fluctuation is different from that due to the classical Kerr nonlinearity.
- When the refractive index change is due to the laser induced tilt angle in smectics we obtain various types of nonlinearities. For instance, in smectic A phase the tilt is induced only beyond a threshold intensity. In case of smectic C the laser field can reduce the tilt and becomes zero at some critical intensity beyond which no further changes are possible. Thus this case leads to saturation nonlinearity.
- The thermal indexing and the suppression of director fluctuations together result in solitons not only with the bell shaped profile but also with a profile similar to that of kinks.
- In case of laser induced suppression of the director fluctuations the critical power

required for the soliton formation is inversely proportional to the cross-section of the beam. This result should be contrasted with that due to the usual Kerr process where the critical power is independent of the beam width. There exists two critical powers for the soliton formation when both this and the thermal indexing are operating simultaneously.

Studies reported in this thesis revealed the richness of the subject of Nonlinear Optics of Liquid Crystals. They have also led to more problems which are being currently pursued. Parts of this thesis have already been published in the following papers:

1. Nematic Kink States in a Laser field, S. K. Srivatsa and G. S. Ranganath, Phy. Rev. E **60(5)**, 5639 (1999)

2: New Nonlinear Optical Processes in Liquid Crystals, S. K. Srivatsa and G. S. Ranganath, Opt. Commun. 180, 349 (2000)

3: Optical Spatial Solitons in Liquid Crystals, S. K. Srivatsa and G. S. Ranganath, 18th International Liquid Crystal Conference, July 2428, 2000, Sendai, Japan.