OPTICS OF TWISTED LIQUID CRYSTALLINE MEDIA

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by

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D E C L A R A T I O N

I hereby declare that the entire work embodied in this thesis is the result of the investigations carried out by me independently in the Liquid Crystal Laboratory, Raman Research Institute, Bangalore, and that no part of it has been submitted for the award of any Degree, Diploma, Associateship or any other similar title.

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CERTIFICATE

I certify that this thesis has been composed by Mr. Yuvaraj Sah based on the investigations carried out by him at the Liquid Crystal Laboratory, Raman Research Institute, Bangalore, under my supervision. The subject matter of this thesis has not previously formed the basis of the award of any Degree, Diploma, Associateship, Fellowship or other similar title.

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This thesis is dedicated to *my* mother and in fond memory of *my father*

Contents

Symbols that are repeatedly used in the thesis

$\hat{n}$	Director
P	Pitch of the twisted liquid crystalline medium
n_1, n_2	Principal values of the index ellipsoid
$\bar{n}$	Mean refractive index
Δn	Birefringence
λ	Wavelength of light
λ_o	Wavelength at the center of the Bragg band
$\Delta\lambda$	Width of the Bragg band
$\epsilon_{i,j}$	Dielectric tensor
$\epsilon_1, \epsilon_3, \epsilon_3$	Principal values of the dielectric tensor
d	Sample thickness
$\overrightarrow{\mathbf{m}}$	Magnetization of the magnetic grains
θ_i	Angle of incidence
ψ	Berreman vector
E_x, E_y	X and Y components of the electric field
H_x, H_y	X and Y components of the magnetic field
$\mathbf{F}$	Propagation matrix
$\mathbf{T_o}$	Transformation matrix for Berreman vector
Φ	Eigenmodes in bounding isotropic media
ρ	Faraday rotatory power
$\mathbf{M}$	Jones M matrix
$\mathbf{N}_o$	Jones N matrix
g_o	Half the phase retardation per unit thickness
k	Wave vector
φ	Azimuth of the incident plane polarized light
q	Scattering vector
T	Temperature
T_c	Smectic A - Sc* transition temperature
0	Tilt angle in the Sc* phase
Ψ	Infinite column vector containing the Fourier components of the transverse fields
$\phi(z)$	Column vector containing the strength of different modes in the isotropic media
$\mathbf{T}$	Transformation matrix for $\Psi(z)$ vectors
S	Scattering matrix
θ_s	Scattering angle
$K_+/\gamma, K_3/\gamma$	Visco-elastic coefficients
τ_G	Relaxation time of the Goldstone mode

PREFACE

This thesis describes some of the theoretical and experimental investigations on the optical properties of chiral liquid crystals namely cholesteric, chiral smectic C (Sc*) and magnetically doped cholesteric phases. The main results are briefly summarized below.

Anomalous transmission in absorbing cholesterics at oblique incidence

One of the interesting effects in the absorbing cholesteric medium is the anomalous transmission. For light propagation along the twist axis of a right handed absorbing cholesteric medium, it is known that the right circular polarized wave is Bragg reflected. Interestingly this Bragg reflected wave is anomalously trarismitted on the shorter wavelength side of the reflection band in comparison to the left circularly polarized wave which only experiences the average absorption of the medium. A similar effect is seen with left circularly polarized light in left handed absorbing cholesteric medium. In chapter 2, we have studied this effect of anomalous transmission at oblique incidence. We consider theoretically light propagation at oblique incidence in absorbing cholesteric liquid crystal in the Bragg mode. We find that the nature of the eigenmodes in the medium changes continuously from the circular to the linear state with increase in angle of incidence. Using appropriate eigenmodes, we have studied theoretically the anomalous transmission in the first and higher order Bragg reflections in these systems and its relevance to reported experimental results. We show that at high angle of incidence ($\theta_i > 55°$) the eigenmodes inside the medium are predominantly linear. At these large angles the Bragg band itself splits into three sub-bands. In the presence of positive linear dichroism we find that the transverse magnetic (TM) wave undergoes anomalous transmission in the short wavelength sub-band. On the other hand with negative linear dichroism, the transverse electric (TE) wave gets anomalously transmitted in the long wavelength sub-band. The similar effect can also be found in the higher order Bragg reflections. Also in the non-Bragg reflection region the reflectance becomes polarization dependent unlike in tlie non-absorbing case where it is unity and is independent of the state of polarization of the incident wave.

Optical properties of magnetically doped cliolesterics

After tlie theoretical work of Brochard and de Gennes on liquid crystal with magnetic grains suspended in them, there have been many studies on different properties of magnetically doped nematic (ferronematic), cholesteric (ferrocholesteric) and smectic (ferrosmectic) phases. It is important to realize that these dispersed grains can not only be transparent but also can exhibit very large inherent Faraday rotation whose optical effect has not been worked out by previous investigators. In chapter **3** we have shown that this inherent Faraday rotation of the magnetic grains can markedly change the optical properties of the ferrocholesterics both in the Bragg mode and in the phase grating mode. In the Bragg mode we consider the case where the grains arc: magnetized along tlie twist axis. We use tlie Jones N matrix to work out the optics of ferrocholesterics in tlie short wavelength side of the reflection band. We find that depending on the propagation of light being parallel or antiparallel to the direction of grain magnetization, the system behaves as a de Vries rotator or as a Mauguin retarder. Further, in the phase grating mode we consider the case where the grains are magnetized perpendicular to the twist axis and the magnetization of the grains rotating with the local director. In this case the optical periodicity of the ferrocholesteric medium is no longer π but 2π rotation of index ellipsoid because of the magnetization of the grains. We have used Raman Nath theory to compute tlie optical diffraction for the linearly polarized light incident normal to the twist axis. Since the optical periodicity is 2π rotation of the index ellipsoid, we get extra orders of diffraction that are not prcsent in normal cliolcstcrics. Wc also find diffraction for any azimuth of the incident light. We have carried out a detailed study of the intensity and the polarization of the different diffraction orders. We bring out the differences between the optical properties exhibited by cholesteric and ferrocholesteric phases.

Optical diffraction in chiral smectic C liquid crystals: Experimental study

When a plane wavefront of polarized light is incident perpendicular to the twist axis of a chiral smectic C (Sc*) liquid crystalline phase, the medium acts as a one dimensional phase grating. The incident wavefront emerges out of the medium with a periodically varying polarization and phase corrugations resulting in the optical diffraction. In chapter 4 we have investigated experimentally the intensity and polarization fcatures of thc optical diffraction in thc phasc grating modc of thc **Sc*** phasc. In our dctailed cxpcriments we find that, for 23, 50 and 125μm thick samplcs, irrespective of the azimuth of the incident light, all the orders are predominantly linearly polarized with azimuth parallel to the twist axis. In the case of 250 μm sample, we get two types of behaviour depending upon the temperature of the sample ; i) At low temperatures the polarization features are same as that found in the thin samples. ii) At higher temperatures there is a reversal of polarization features, i.e., the diffracted light is nearly linearly polarized with azimuth perpendicular to the twist axis. The intensity and polarization features of the diffracted light are not in agreement with the theoretical results predicted by the simple Raman-Nath theory for the Sc* phase.

Optical diffraction in chiral smectic C liquid crystals: Theoretical study

To account for the experimental results discussed in the previous chapter, we have used a theory based on the Rokoshima and Yamakita (RY) approach for anisotropic dielectric gratings to compute the diffraction pattern for Sc* in the phase grating mode. This is the subject matter of chapter 5. This theory takes care of the internal diffractions inside the sample. It is found that the intensity and polarization of the diffraction ordcrs are sensitive to tlie sample thickness and rnatcrial parameters, i.e., tilt angle, pitch and birefringence. All these parameters arc temperature dependent in the Sc* phase. We find the experimental results can be accounted for by this rigorous tlicory. In addition, one ol the interesting results ol these exact calculations is the oscillations in the intensity of different diffraction ordcrs with sample thickness. Using a perturbation theory wc interpret tlie modulations as arising from a coupling between different ordcrs of scattering of tlie diffracted light.

Optical diffraction in a quasi-periodic liquid crystalline media

Cholesteric liquid crystalline phases with pitch ≤ 1 μm invariably go through blue phases before melting into the isotropic phase. Blue phases (BP) are of three types.

131' I and BP II have been structurally characterized. But the structure of BP III is not yet very well established. Interestingly a particular model of BP III has quasi-periodic ordering. In view of this model, in chapter 6, we consider a one dimensional quasi-periodic cholesteric. This is a quasi-periodic medium which is optically anisotropic in nature. We have addressed ourselves to a Fibonacci sequence of helically stacked birefringent layers. The optical properties of such a medium can give an insight into the structure of the BP III. For the light propagation in the Bragg mode through such a quasi-periodic structure, we find that the reflection spectrum comprises of many reflection bands. The number of Bragg bands increases with the sample thickness. Each band behaves like a normal cholesteric band. We find self-similarity in the reflection spectra. Also the linear dichrosim suppresses various reflection bands. In the phase grating mode, unlike the normal cholesteric phase, we find that the diffraction spectra contain a large number of diffraction orders. The position of the intense diffraction peak can be assigned by a pair of integers. Interestingly these two integers are not successive Fibonacci numbers as found in Fibonacci amplitude gratings.

During our studies on the optical diffraction on Sc*, we have also investigated this phase using dynamic light scattering. With this technique we liavc calculated the visco-elastic coefficients of the compound in the Sc* phase. Apart from the Goldstone mode we observe a relatively slow varying mode in the Sc* phase for the sample aligned in the phase grating geometry. This new mode is nearly independent of the scattering vector unlike the Goldstone mode which has a quadratic dependence on the scattering vector. Appendix A carries a brief presentation of this work.

Apart from our study on optical diffraction in a quasi-periodic cholesteric medium, we have also considered diffraction in some more Fibonacci structures. Appendix B contains a summary of the results of this study.

The main results of this thesis work have been reported in the following publications:

1. Yuvaraj Sah, K.A. Suresh and G.S. Ranganath, "Effect of inherent Faraday rotation on the optical properties of *Ferrocholesterics*", presented in the 14th international liquid crystal conference June, 1992 held in Pisa (Italy).

2. Yuvaraj Sah, K.A. Suresh and G.S. Ranganath, "Optical properties of *magnetically* doped *cholesterics*", Liquid Crystals, 15, 25 (1993).

3. Yuvaraj Sah and K.A. Suresh, "*Anomalous transmission at oblique incidence* in absorbing *cholesteric* liquid crystals", J. Opt. *Soc. Ame.* A, **11**, 740 (1994)

4. K.A. Suresh, Yuvaraj Sah, P.B. Sunil Kumar and G.S. Ranganath, "Optical diffraction in chiral *smectic* C *liquid* crystal", Phys. Rev. Lett., 72, 2863 (1994).

5. Yuvaraj Sah and G.S. Ranganath, "Optical diffraction in some Fibonacci *structures*", Optics Communications, 114, 18 (1995).

6. Yuvaraj Sah, P.B. Sunil Kumar and K.A. Suresh, "Intensity and *polarization* features in the optical diffraction in *chiral* smectic C liquid crystals", Sent for publication.