Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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To cite this article: K. A. Suresh (1997): Optics of Some Chiral Liquid Crystals, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 302:1, 235-246

To link to this article: http://dx.doi.org/10.1080/10587259708041834

Mol. Cryst. Liq. Cryst., 1997, Vol. 302, pp. 235–246
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OPTICS OF SOME CHIRAL LIQUID CRYSTALS

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Abstract Although the optics of chiral systems like cholesterics has been studied for well over one hundred years, the subject is still very relevant in view of new and different chiral systems that have been realised in the laboratory in recent times. These systems are associated with novel optical properties and have drawn considerable attention. Here, we consider the optics of ferrocholesterics, absorbing cholesterics, chiral smectic C, absorbing and non-absorbing twist grain boundary smectics and a tapered soliton lattice. In particular, we deal with reflection, transmission and diffraction properties of these systems.

INTRODUCTION

The well known example of a chiral liquid crystal is a cholesteric liquid crystal which exhibits interesting optical properties like Bragg reflection, very high rotatory power and its anomalous dispersion. The other examples are chiral smectic C, ferrocholesterics, twist grain boundary smectics (TGBS) and twist soliton lattices. Here we will be concerned with these systems.

We will highlight some of the recent interesting results that have been obtained by us viz., 1) Asymmetric transmission along the twist axis in ferrocholesterics. 2) Polarisation sensitive non-Bragg reflections in absorbing cholesterics. 3) Modulations in the diffracted intensity in chiral smectic C. 4) Unusual reflection spectra in TGBS and finally (5) asymmetric reflections in a tapered soliton lattice.

FERROCHOLESTERICS

Cholesteric phases of rod-shaped molecules in which needle shaped magnetic grains are aligned along the local director have been realized in the laboratory. There are systems with grains having magnetization parallel to the local director[1] as well as magnetization perpendicular to the local director[2]. The former will give rise to a magnetically doped cholesteric (ferrocholesteric) phase with magnetization gradually twisting with the local director much like a helimagnetic system and in the latter case we can get the same phase with the magnetization of the grains parallel to the twist axis. Since these grains can even be optically transparent (as in the case of grains of garnets), their inherent Faraday rotation becomes important.

The rotatory power due to magnetic grains depends on the direction of propagation of light with respect to the magnetization m and is given by

$$\bar{\rho} = F \mid \mathbf{m} \mid \cos\Omega = \bar{\rho}_o \cos\Omega$$

where F is a constant and Ω the angle between m and the direction of propagation. This dependence of the Faraday rotation on Ω leads to optical properties which are very different[3] from those of the classical cholesterics. An interesting case, with light propagation parallel to the twist axis is discussed here.

Consider the system where the magnetic grains are parallel to the local director but with m along the twist axis. The medium at any point acts as a linearly birefringent plate with Faraday rotation.

We use the Jones matrix formulation to work out the optics in the limit of the wavelength being much smaller than the optical period of the structure, i.e., ignoring multiple reflections. The N matrix for such a plate is given by[4]

$$\mathbf{N}_o = \begin{bmatrix} -ik + i\gamma & -\bar{\rho} \\ \bar{\rho} & -ik - i\gamma \end{bmatrix},$$

where k is the wavevector in the medium and $\gamma = \pi\Delta n/\lambda$, with Δn being the layer birefringence of the medium, λ, the wavelength of light and $\bar{\rho}$ is the Faraday rotatory power. We consider the propagation along the z axis. Then the N matrix of a layer at z is given by

$$\mathbf{N} = S(\beta z)\mathbf{N}_o S(-\beta z).$$

Here S(βz) is a rotation matrix with $\beta = 2\pi/P$, P being the pitch. The Jones matrix M for the entire sample can then be written as

$$\mathbf{M} = S(\beta z)\exp[\{\mathbf{N}_o - \beta S(\pi/2)\}z].$$

Case 1

$\mid \bar{\rho} - \beta \mid \gg \mid \gamma \mid$ with $\bar{\rho}$ and β of opposite signs i.e., when the direction of propagation of light is opposite to that of m. Then,

$$\mathbf{M} = \exp(ikz)S(\bar{\rho}z)$$

Hence the medium, to a very good approximation, acts as a pure rotator, i.e., as in the de Vries limit.[5] This condition can also be satisfied for $\beta \ll \gamma$. Thus the existence of Faraday rotation can lead to a de Vries limit even for a medium of large pitch. This is contrary to the case of classical cholesterics where the de Vries limit is reached only at very small pitches.

Case 2

$|(\bar{\rho}-\beta)| \ll |\gamma|$ with $\bar{\rho}$ and β of the same sign (i.e., when the propagation of light is in the same direction as that of $\mathbf{m}$). In this case we find,

$$\mathbf{M} = \exp(ikz)\begin{bmatrix} \cos\beta z & -\sin\beta z \\ \sin\beta z & \cos\beta z \end{bmatrix}\begin{bmatrix} \exp(i\gamma z) & 0 \\ 0 & \exp(-i\gamma z) \end{bmatrix}$$

This describes the Mauguin limit.[6] This condition can be satisfied even when $\beta \gg \gamma$. Hence due to Faraday rotation the solution will go over to Mauguin limit, instead of de Vries limit.

Therefore depending on whether the propagation is along or opposite to the direction of $\mathbf{m}$ the medium acts as a Mauguin retarder or as a de Vries rotator, respectively. For example, this happens in a ferrocholesteric of pitch $\simeq 30\mu$m, for $\bar{\rho}_o \simeq 2.0\times10^3$ rad cm^{-1} and birefringence $\Delta n = 0.025$. Such a medium between two appropriately aligned polaroids can act as an optical diode, i.e., transmitting light in one direction and blocking it completely in the opposite direction.

ABSORBING CHOLESTERICS

In X-rays, Borrmann[7] discovered that there is an enhancement in the intensity of transmission when a perfect absorbing crystal is set for Bragg reflection. A similar phenomenon has been predicted in absorbing cholesterics.[8] The speciality of the latter case is that the polarization of the standing wave inside the medium and the linear dichroism of the molecules play a significant role. The physical origin of this effect in cholesterics may be explained as follows. Consider a right circularly polarized light incident on a right handed cholesteric planar film. In the reflection band, a standing linear wave is set up inside the medium due to the interference between the forward and backward propagating right circularly polarized waves. The net electric vector (E) of the standing wave in the medium is orthogonal to the director on the shorter wavelength side and is along the director on the longer wavelength side and the azimuth of the linear standing wave follows the director. Generally the linear birefringence and linear dichroism of the molecules are positive. In such cases the net linear vibration experiences minimum and maximum absorption respectively at these two edges of the reflection band. As a result there occurs an anomalous increase of the transmitted intensity on the shorter wavelength side compared to the transmitted intensity of the non-reflected left circular wave that experiences an average absorption in the medium. That such an enhancement in the transmitted intensity occurs has been demonstrated experimentally.[9,10]

An effect related to the anomalous transmission has been studied experimentally at oblique incidence by Endo *et al.*[11] They found that the anomalous transmission occurs only at angles of incidence, less than 19°. This has been accounted for in a recent theoretical study[12] on light propagation at oblique incidence. The computations show that the eigenmodes, change gradually from circular to linear state as one increases the angle of incidence. In this theory we have employed Berreman's[13] 4 × 4 matrix theory of light propagation at oblique incidence. It can be shown[12] that

$$\begin{bmatrix} t_1 \\ t_2 \end{bmatrix} = D_1 \begin{bmatrix} i_1 \\ i_2 \end{bmatrix} \quad \text{and} \quad \begin{bmatrix} r_1 \\ r_2 \end{bmatrix} = D_2 \begin{bmatrix} i_1 \\ i_2 \end{bmatrix}$$

Here i_1, i_2 are the incident, r_1, r_2 are the reflected and t_1, t_2 are the transmitted amplitudes of the respective transverse magnetic (TM) and transverse electric (TE) polarizations (with respect to the plane of incidence) inside the bounding isotropic medium. The eigenvectors of the transmission and reflection matrices D_1 and D_2 represent the forward and the backward propagating eigenmodes respectively, in the isotropic medium. These eigenmodes in the isotropic medium bounding the cholesteric are responsible for forming the standing and the propagating waves inside the cholesteric medium. For example, at normal incidence the eigenvectors of D_1 and D_2 are the orthogonal circular waves. The resultant vibration of one of these waves gives rise to a linearly polarized standing wave with **E** parallel to **H** and the other is a propagating circular wave.[14]

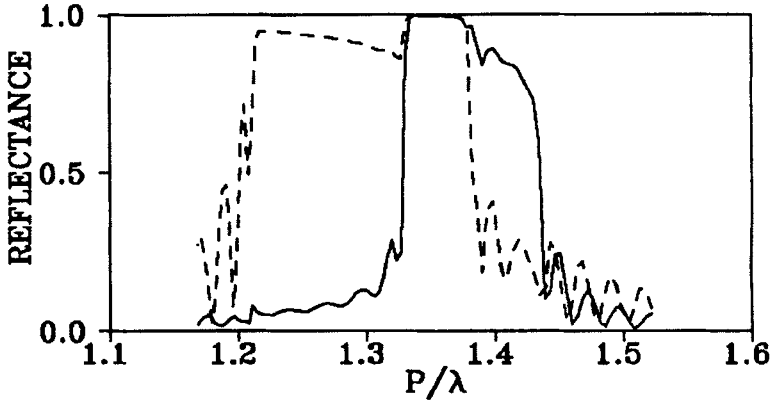

FIGURE 1 The reflectance for the TM (continuous curve) and TE (long-dashed curve) waves for a non-absorbing cholesterics at $\bar{\theta} = 60°$.

At large angles of incidence ($\bar{\theta} > 58°$) the eigenvectors of D_1 and D_2 matrices are predominantly linear and also the nature of the eigenmodes does not change within the reflection band. At these large angles for a non-absorbing cholesteric the Bragg band splits into three sub-bands. In the short wavelength sub-band the TM wave is reflected considerably and the TE wave is almost fully transmitted. In the long wavelength sub-band it is the TE wave which is strongly reflected and the TM wave is almost fully transmitted. The central sub-band is a non-Bragg reflection band inside which both TE and TM waves are totally reflected. This is shown in figure 1. At large angles the nature of the eigenmodes do not change with the wavelength throughout the sub-bands. In the absorbing cholesteric medium one can use the TE and TM waves which are the eigenvectors of D_1 and D_2 to study the anomalous transmission. The propagation of the TE and TM waves in the absorbing cholesterics is such that in the short wavelength region the TM wave which is reflected more also shows an enhanced transmission. This is shown in Figures 2a and 2b.

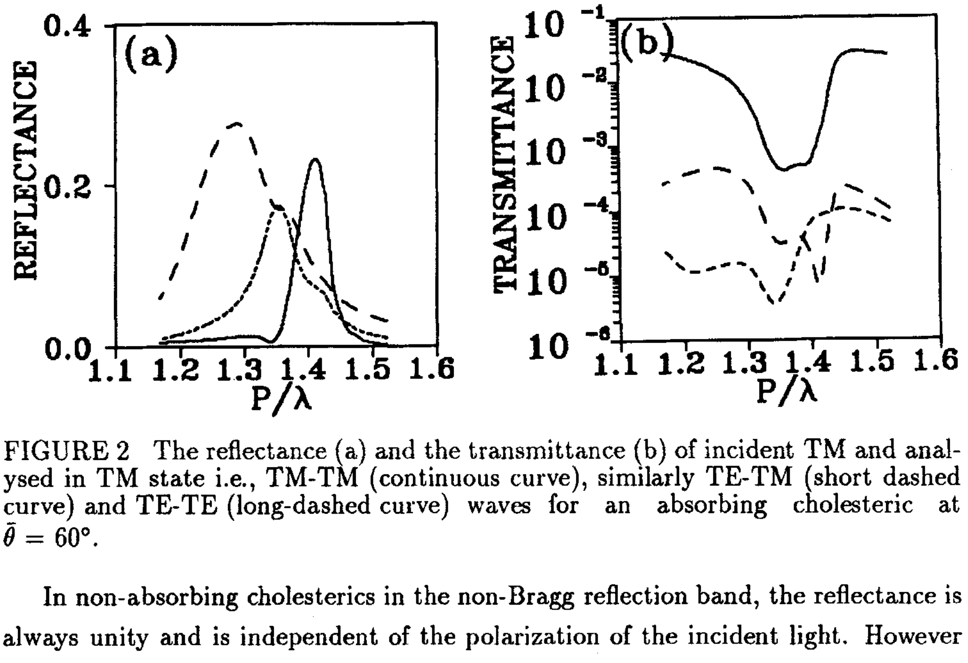

FIGURE 2 The reflectance (a) and the transmittance (b) of incident TM and analysed in TM state i.e., TM-TM (continuous curve), similarly TE-TM (short dashed curve) and TE-TE (long-dashed curve) waves for an absorbing cholesteric at $\bar{\theta} = 60^\circ$.

In non-absorbing cholesterics in the non-Bragg reflection band, the reflectance is always unity and is independent of the polarization of the incident light. However the theory predicts[12,15] that in the absorbing case, the reflectivity in this band is very much dependent on the state of polarization of the incident light. This dependence of the reflectance on polarization can be understood by considering the intensity of the electric field of the non-propagating eigenmodes worked out from Berreman's 4×4 matrices. In the absence of absorption the fields of both the non-propagating eigenmodes suffer attenuation to the same extent. But in the presence of absorption the two eigenmodes suffer different amounts of absorption and therefore one of the eigenmodes gets attenuated more. Thus the reflectance becomes polarization dependent.

CHIRAL SMECTIC C

The chiral smectic C (Sc*) liquid crystal is a helical stack of layers of uniformly tilted molecules. The tilt of the molecule coupled with constant layer thickness produces local biaxiality in the medium. Here, for the propagation of light perpendicular to the twist axis , the medium acts as a phase grating.

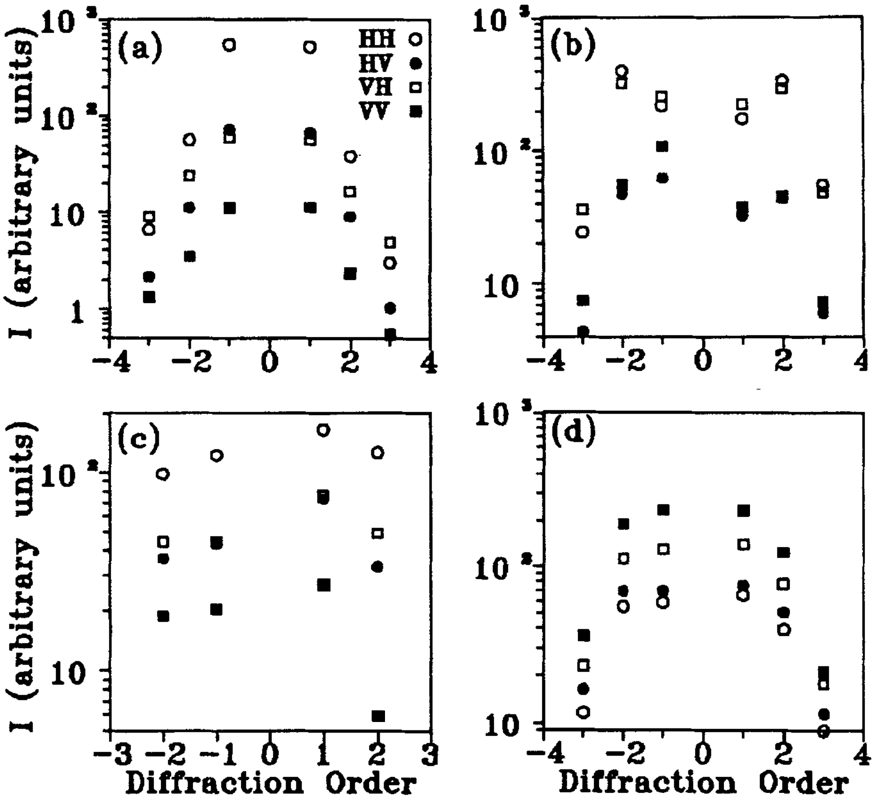

FIGURE 3 The measured intensity I (in arbitrary units) of the diffraction orders for various geometries,(a) for d=23 μm and temperature t=50.6° C, (b) for d=125 μm and t = 49.3 ° C (c) for d=250 μm and t = 45.5 °C. (d) for d=250 μm and t = 50.6 °C. (Since the intensity of the zeroth order in all the cases is too high, it is not indicated in the figure.)

Recently we studied[16,17] diffraction in the phase grating mode in a BDH-SCE6 liquid crystal. The SCE6 material on cooling from the isotropic phase exhibits successively cholesteric (120°C) smectic A (81°C) Sc* (63°C) and crystalline ($\leq$ 15°C) phases. In the experiment, polarized light from a He-Ne laser(λ = 0.6328μm)was sent in a direction normal to the twist axis of Sc* resulting in a diffraction pattern.

The diffracted intensity was analysed in the HH, HV, VV and VH polarization geometries.(H denotes polarization parallel to the twist axis and V denotes polarization perpendicular to the twist axis. The first symbol indicates the state of polarization of the incident light and the second symbol indicates the polarization state in which the diffracted beam is analysed).

The measured intensities of different diffraction orders are depicted in figures 3a and 3b for sample thicknesses 23 and 125 μm respectively in HH, HV, VH and VV geometries. One finds that the intensity in the HH geometry is always higher than the intensity in the HV geometry for all orders. Also the intensity in the VH geometry is always higher than that of the intensity in the VV geometry. In other words, in all the orders, the diffracted light is nearly linearly polarized parallel to the twist axis. This behaviour is observed in samples of thickness 23 and 50 μm at all temperatures in the Sc* phase. At lower temperatures, away from the Sc* - S_A transition point, the 125 μm thick sample also exhibits the same behaviour. For the 250 μm thick sample one finds that at low temperatures, the intensity and polarization features are similar to those found in thinner samples (figure 3c). But at higher temperatures, the behaviour is completely reversed i.e, the intensity in the VV geometry becomes more than the intensity in the VH geometry (figure 3d). Even the intensity in the HV geometry becomes more than that in the HH geometry, i.e., in this case, in all the orders, the diffracted light is nearly polarized perpendicular to the twist axis. Thus in this thicker sample, one can drastically change the polarization features of the diffraction orders by varying the temperature. In all these samples, the zeroth order has nearly the same state of polarization as that of the incident light.

The problem of optical diffraction in Sc* can be theoretically formulated in two ways: i) Generalization of Raman-Nath theory of phase gratings [18]. ii) Extension of Rokushima, Yamakita [19] and Galatola et. al. [20] (RY) theories of anisotropic dielectric gratings. The first approach is quite physical and leads to a simple but approximate answers, since this ignores the internal diffraction in the medium. Hence it is applicable to a medium of low birefringence and of small sample thicknesses. The second approach is a rigorous theory and is applicable to even highly birefringent and thick samples. Also the RY theory leads to the same results as that of the Raman-Nath theory in the limit of low material birefringence and small sample thicknesses. Here we present the generalised RY theory.

RY theory

We consider S_C^* with its twist axis along the y direction and light propagation along the z direction. To work out the optics of the anisotropic dielectric media one writes the Maxwell equations in the form [19,20]

$$\frac{d\Psi(z)}{dz} = ik_0 D\Psi(z)$$

where,

$$\Psi(z) = \begin{bmatrix} e_x \\ h_y \\ e_y \\ h_x \end{bmatrix}$$

Here e_x, e_y, h_x and h_y are sub-matrices of the infinite column matrix $\Psi(z)$ at any point z and contain the various Fourier components of the transverse field. Here $k_0 = 2\pi/\lambda$. The propagation matrix D is an infinite square matrix.[16]

From the modal analysis,[20] we find

$$\phi_r = \mathcal{R}\phi_i \ , \ \phi_t = \mathcal{T}\phi_i$$

where ϕ_i, ϕ_r and ϕ_t are the incident, reflected and transmitted components respectively of the column vector containing the strengths of different modes in the isotropic medium. $\mathcal{R}$ and $\mathcal{T}$ are the reflection and transmission matrices respectively. The RY theory can qualitatively account for the experimental results.[16]

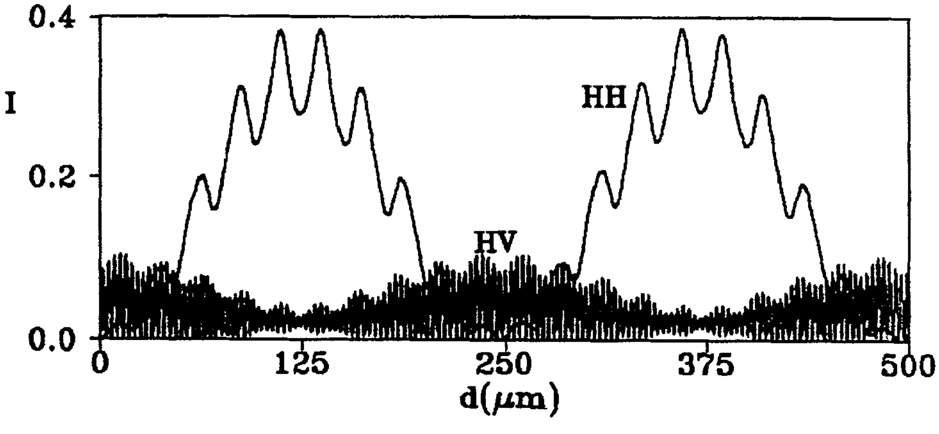

FIGURE 4 Normalised intensity I for the first order diffraction as a function of sample thickness for the HH and HV geometries. The parameters used in the calculation are; pitch P = 5 μm, birefringence Δn=0.18 and tilt angle θ=18°.

Modulations in the diffracted intensity

We have computed the diffraction intensity using the transmission matrix $\mathcal{T}$. The first order diffraction intensity in two different geometries is given in figure 4. One can notice that, in the HV geometry, the diffracted intensity has fine fringes of width 4 μm along with modulations at about 25 and 250μm. For the HH geometry, the modulations are 250 and 25 μm. One gets such modulations in the intensity in

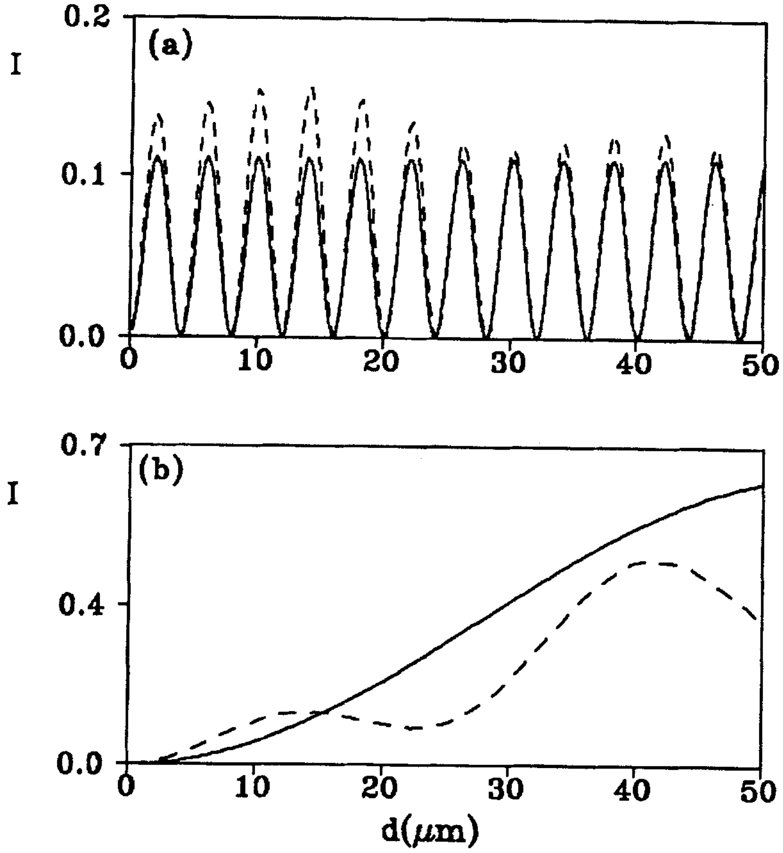

FIGURE 5. The normalized intensity I of the first order diffraction as a function of sample thickness computed using a perturbation theory (a) for the HV geometry, (b) for the HH geometry. The full line represents the contribution from the first order perturbation and the dashed line represents third order perturbation contribution to the diffracted intensity.

the other geometries also. Such effects are also present in the second order. We use a perturbation technique [20] to account[21] for these modulations. This technique is a simpler but approximate method of computing the diffracted intensity from the scattering matrix S. In this procedure we consider at the propagation equation for the S matrix i.e.,

$$\frac{dS}{dz} = i\ k_0 G S$$

The matrix G can be written as a sum of its diagonal matrix G_o and an off-diagonal matrix g. G_o contains the strength of the zeroth order Fourier components of the dielectric tensor of an effective homogeneous anisotropic medium. The matrix

g contains the strength of the higher order Fourier components which arises due to the z dependence part of the dielectric tensor. It may be noticed that mathematically, the above equation is rather analogous to the time dependent Schrödinger equation [22] Based on this analogy we use [21] the time dependent perturbation method of quantum mechanics[22] to solve this problem.

The matrix g can be treated as a perturbation over the matrix G_o. Then we can work out the first, second and third order scattering contributions to the amplitude of the diffracted light, viz., $(A_{S_I})_{\ell j}$, $(A_{S_{II}})_{\ell j}$ and $(A_{S_{III}})_{\ell j}$. Figure 5a shows the contributions of different orders $(A_{S_I}, A_{S_{II}}, A_{S_{III}} \ldots)$ of perturbations to the intensity of the first order diffraction. Further, we find a coupling between zeroth order diffracted TM and first order diffracted TE polarizations in HV geometry. We see that the first order perturbation $(|\,(A_{S_I})_{0_H 1_V}\,|^2)$ only results in the fine modulations at 4μm. The third order perturbation $(|\,(A_{S_I})_{0_H 1_V} + (A_{S_{II}})_{0_H 1_V} + (A_{S_{III}})_{0_H 1_V}\,|^2)$ leads to modulation with a larger period of 25 μm. Figure 5b shows the first $(|\,(A_{S_I})_{0_H 1_H}\,|^2)$ and third $(|\,(A_{S_I})_{0_H 1_H} + (A_{S_{II}})_{0_H 1_H} + (A_{S_{III}})_{0_H 1_H}\,|^2)$ order perturbation contributions to the intensity in the first order diffraction. In this case, the coupling is between the zeroth order TM polarization and the first order TM polarization in HH geometry. Here we find that the first order perturbation contribution corresponds to the modulation of 250 μm period and the third order perturbation has, in addition, the modulation of 25 μm period seen in figure 4 in the HH geometry. For the parameters that we used, the contributions due to second order perturbation is not very different from that due to the first order perturbation.

We would like to mention that the perturbation technique does not incorporate the effect of reflectance. However, we find from the rigorous theory that the reflectance effects are negligible.[21] Also the boundary effects do not alter the scale of the modulations present in the diffracted intensity. It may be remarked that the perturbation calculations are valid only for thin samples. However, this technique gives a qualitative understanding of the modulations in the diffracted intensity obtained from the rigorous theory.

TWIST GRAIN BOUNDARY SMECTICS

Twist grain boundary smectic phases are endowed with many interesting properties. In this phase, thick smectic blocks are helically stacked with intermediate cholesteric like twist grain boundaries. The important feature of the structure is the possibility of incommensuration between the pitch of the helix and the thickness of the smectic blocks. Optically the smectic blocks can be thought of as thick birefringent plates which are arranged in the helical stack. Any two such blocks are connected by a grain boundary which can be thought of as a helical stack of thin birefringent plates.

On this model, we have worked out[23] reflection and diffraction properties. Here, we highlight some of the interesting results obtained by us. In the case of non-absorbing systems, at normal incidence, one gets multiple reflections. Interestingly, irrespective of the handedness of the structure one finds[24] bands which reflects strongly either left circular or right circular incident light. One also finds some bands that reflect any state of polarised light. When the system is absorbing, we find that bands associated with circular state exhibit Borrmann transmission. The characteristics of this transmission are very sensitive to the uniformity of the optical absorption. That is, whether or not the absorption is the same in both the smectic blocks and in the grain boundaries. Generally, the anomalous transmission is invariably in the shorter wavelength side of the reflection band though there could be some bands exhibiting this even at the longer wavelength side. Surprisingly, there could be some bands which do not at all exhibit Borrmann effect. Further, in the case of absorption, confined only to the grain boundary, we find[23] absence of the Borrmann effect in some bands which under uniform absorption exhibited this effect.

TWIST TAPERED SOLITON LATTICES

Generally, a planar twist soliton connects a uniform director orientation to equivalent uniform state through a twist wall. However, under certain dynamical situations, we can find a planar twist soliton which connects a uniform director orientation state to one with a periodic director distortion with the amplitude of the director distortion decaying exponentially. We call them twist tapered solitons. Its optics has also been worked out.[23] We recall here its unusual reflection properties. Again for light propagation parallel to the twist direction, we find a multiplicity of bands. Interestingly, each band strongly reflects both right and left circularly polarised light but with slightly different strengths. Further, the reflection co-efficient is very different for light propagation along or opposite to the twist direction. In these systems, optical absorption leads to an enhancement of the above effects.

ACKNOWLEDGEMENT

I am thankful to G.S. Ranganath, A.Bhattacharyya and N.Andal for helpful discussions.

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