Chapter 2

Anomalous transmission in absorbing cholesterics at oblique incidence

2.1 Introduction

This chapter deals with some theoretical studies on the light propagation at oblique incidence in absorbing cholesterics in the Bragg mode. Cholesteric mediurn can be made anisotropically absorbing (i) if it is composed of molecules which are dichroic, (ii) if the absorption band of the constituent molecules is in the neighbourhood of its Bragg band or (iii) by doping it with a small amount (**1** to **5 %**) of suitable dye having linear dichroism. The dye molecules get dispersed uniformly in the cholesteric medium making it locally linear dichroic [1]. The anomalous transmission is the most interesting phenomenon found in absorbing cholesterics. For a right handed cholesteric medium, in the absence of absorption, right circularly polarized (RC) wave gets Bragg reflected and is transmitted less as compared to left circularly polarized (LC) wave throughout the reflection band. But surprisingly when the linear dichroism is considered the RC wave gets more transmitted than the LC wave on the shorter wavelength side of the reflection band. This anomalous transmission is shown in figure **2.1 .** As stated earlier this anomalous transmission is the optical analogue of the Borremann effect in X-rays [2]. At normal incidence it has been well studied theoretically and experimentally [3-6].

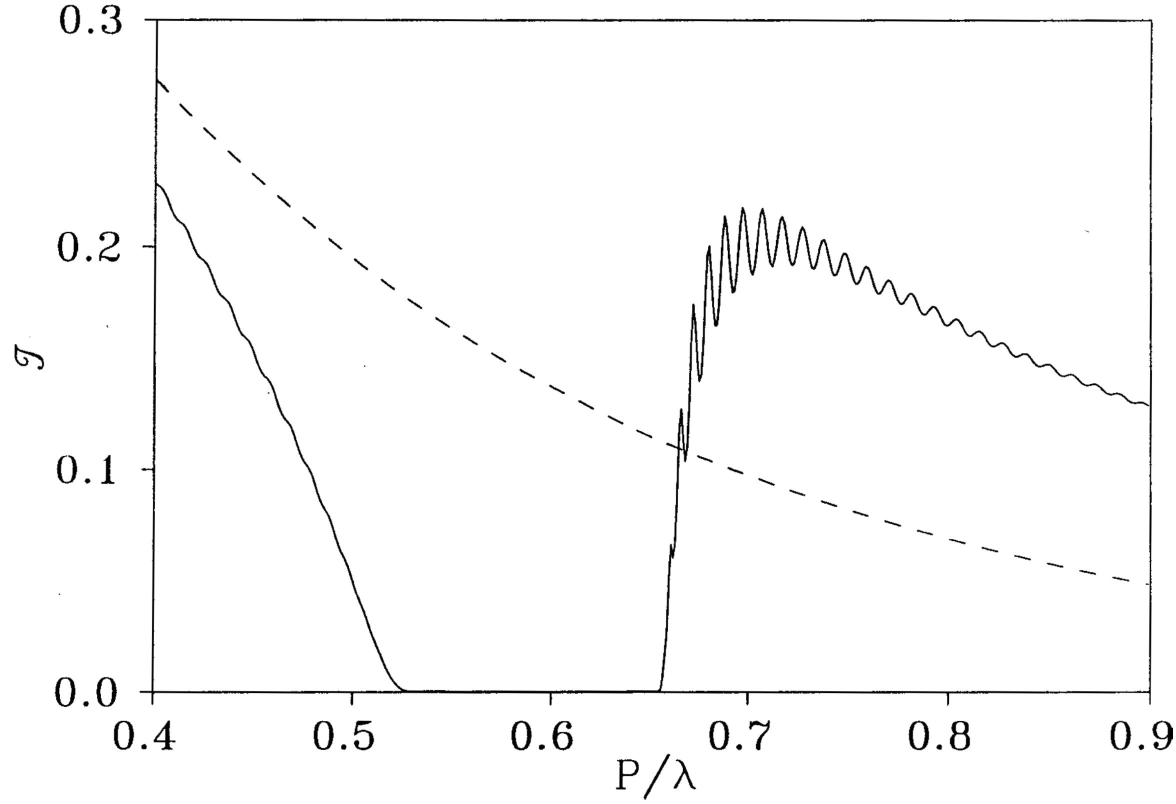

Figure 2.1: The transmittance of the RC (solid line) and LC (dashed line) waves at normal incidence through an absorbing cholesteric.

The reason for this anomalous transmission can be understood by appealing to the standing waves built inside the medium in the reflection band. It has been shown [6] that the standing waves inside the medium are locally linear in nature. The azimuth of this linear vibration rotates along with the director as the wave traverses inside the medium. Also the azimuth of this vibration rotates by $\pi/2$ as we move from one edge of the reflection band to other edge. If we assume that the linear dichrosim is positive, i.e., the absorption is more for this vibration along the long axis of the molecules compared to that in the perpendicular direction then the RC wave will experience less absorption at short wavelength edge of the reflection band and gets anomalously transmitted while LC wave suffers uniform absorption throughout the reflection band. If we consider the linear dichroism to be negative, i.e, the absorption is more along the short axis of the molecules compared to that in the long axis then this anomalous transmission shifts to the long wavelength edge of the reflection band. In this chapter we have looked into this phenomenon when light is incident at an angle to the twist axis (oblique incidence).

2.2 Theory

The analytical treatment of the problem of light propagation in cholesteric for arbitrary angle is quite complicated. However, the problem has been solved rigorously for normal incidence [6]. In the case of oblique incidence it has been solved analytically only within some approximations [7-9]. But it is rather straightforward to solve the problem numerically using Berreman's 4 x 4 matrix formulatiori [10]. Here we use this method to solve the Maxwell's equations numerically. In this formulation the Maxwell's equations can be written as

$$\frac{\partial \psi}{\partial z} = \frac{iw}{c}\Delta(z)\psi \tag{2.1}$$

where

$$\psi = \begin{bmatrix} E_x \\ H_y \\ E_y \\ -H_x \end{bmatrix}$$

and

$$\Delta(z) = \begin{bmatrix} -m\frac{\epsilon_{xz}}{\epsilon_{zz}} & 1-\frac{m^2}{\epsilon_{zz}} & -m\frac{\epsilon_{yz}}{\epsilon_{zz}} & 0 \\ \epsilon_{xx}-\frac{\epsilon_{xz}^2}{\epsilon_{zz}} & -m\frac{\epsilon_{xz}}{\epsilon_{zz}} & \epsilon_{xy}-\frac{\epsilon_{xz}\epsilon_{yz}}{\epsilon_{zz}} & 0 \\ 0 & 0 & 0 & 1 \\ \epsilon_{xy}-\frac{\epsilon_{xz}\epsilon_{yz}}{\epsilon_{zz}} & 1-\frac{m^2}{\epsilon_{zz}} & \epsilon_{yy}-\frac{\epsilon_{yz}^2}{\epsilon_{zz}}-m^2 & 0 \end{bmatrix}$$

Here E_x, E_y and H_x, H_y are the X and Y components of the electric and magnetic fields respectively, ϵ_{xy} etc. are the components of the dielectric tensor and m = $\bar{n}sin(\theta_i)$, where 8; being the angle of incidence. Twist axis is chosen to be in Z direction.

We use the Oseen's model to get the local dielectric tensor for the cholesteric medium. In this model, the dielectric tensor is given as follows

$$\epsilon_{ij} = \begin{pmatrix} \bar{\epsilon}+\delta\cos(2\alpha) & \delta\sin(2\alpha) & 0 \\ \delta\sin(2\alpha) & \bar{\epsilon}-\delta\cos(2\alpha) & 0 \\ 0 & 0 & \epsilon_2 \end{pmatrix} \tag{2.2}$$

$\bar{\epsilon} = (\epsilon_1 + \epsilon_2)/2$, $\delta = (\epsilon_1 - \epsilon_2)/2$ and $\alpha = (2\pi/P)$z, where P is the pitch of the medium, ϵ_1 and ϵ_2 are the principal values of the local dielectric tensor. In our calculations we have chosen $\epsilon_1 = 2.3228$ and $\epsilon_2 = 2.5673$ for the non-absorbing cholesteric medium and $\epsilon_1 = 2.3228 + i$ x 0.0063 and $\epsilon_2 = 2.5673 + i \times 0.063$ for the absorbing cholesteric medium.

The equation (2.1) can be integrated numerically for one pitch to get the propagation matrix $\mathbf{F}(z,P)$ for the cholesteric medium such that $\mathbf{F}(z,P)$ relates the field at point $(z+\mathrm{P})$ to the field at z given by

$$\psi(\ z+P\ ) = \mathbf{F}(\ z,P\ )\ \psi(\ z\ ) \tag{2.3}$$

The eigenvectors ζ_i (where i = 1,2,3 and 4) of $\mathbf{F}(z,\mathrm{P})$ correspond to the four eigenmodes in the medium and its eigenvalues are related to the wavevectors of these eigenmodes. For a non-absorbing medium, the eigenvalue of a mode will be real if it is a propagating mode and will be complex if the mode is non-propagating. In other words, mode with complex eigenvalues represents the standing wave inside the medium. The components of a particular eigenvector at any point z_1 situated at an infinitesimal distance h from $z=0$ can be obtained by

$$\zeta_i(z_1) = \exp\left(\frac{iw}{c}\Delta\ h\right)\ \zeta_i(0) \tag{2.4}$$

In computing the total intensity of the E field for a particular mode inside the medium for the obliquely incident light we also have to add the Z component of the E field.

In an experimental situation, the cholesteric medium is sandwiched between two glass plates (isotropic medium). It is then convenient to analyze the transmitted and the reflected waves in terms of the forward and backward propagating modes inside this bounding isotropic medium. To do this we use a transformation [11]

$$\psi = \mathbf{T_o}\ \Phi \tag{2.5}$$

Here, ψ is the Berreman vector, $\mathbf{T}_o$ is a 4x4 matrix with columns representing the eigenvectors of the eigenmodes in the bounding isotropic medium and

$$\Phi = \begin{bmatrix} t_{TM} \\ t_{TE} \\ r_{TM} \\ r_{TE} \end{bmatrix}$$

t_{TM} and t_{TE} represent complex amplitudes of transverse magnetic (TM) and transverse electric (TE) modes propagating in the forward direction arid r_{TM} and r_{TE} are the amplitudes for the same modes in the backward direction in the isotropic medium. (For the TM mode the electric vector lies in the plane of incidence and for TE mode it is perpendicular to the plane of incidence.)

We consider a cholesteric liquid crystal aligned in the Bragg mode bounded with two glass plates situated at $Z = 0$ and $Z = nP$. Here n represents the number of cholesteric pitches. Then by arranging the columns in $\mathbf{T}_o$ we can write at the interface $Z = 0$

$$\psi(0) = \mathbf{T_o}\ \Phi_I(0) \tag{2.6}$$

where

$$\Phi_I(z) = \begin{bmatrix} i_1 \\ i_2 \\ r_1 \\ r_2 \end{bmatrix}$$

and

At the second interface $Z = nP$

$$\psi(\ nP\ ) = \mathbf{T_o}\ \Phi_I(\ nP\ ) \tag{2.7}$$

where

$$\Phi_I(\ nP\ ) = \begin{bmatrix} t_1 \\ t_2 \\ 0 \\ 0 \end{bmatrix}$$

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 TM and TE waves inside the isotropic medium. The Berreman vector $\psi(0)$ at $Z = 0$ is related to $\psi(nP)$ by the propagation matrix for n pitches of the cholesteric medium $\mathbf{F}(\ z, nP\ )$

$$\psi(\ 0\ ) = \mathbf{F}^{-1}(\ z, nP\ )\ \psi(\ nP\ ) \tag{2.8}$$

Propagation matrix for n pitches, $\mathbf{F}(\ z, nP\ )$ is related to the matrix $\mathbf{F}(z, \mathbf{P})$ by the relation $(\mathbf{F}(z, nP) = (\mathbf{F}(z, P))^n$. Substituting for $\psi(0)$ and $\psi(nP)$ from equation (2.6), (2.7) to (2.8) we get

$$\Phi_I(0) = C\ \Phi_I(\ nP\ ) \tag{2.9}$$

where $\mathbf{C} = \mathbf{T_o}^{-1}\ \mathbf{F}^{-1}\ \mathbf{T_o}$.

Equation (2.9) could be easily be split into two parts

$$\begin{pmatrix} t_1 \\ t_2 \end{pmatrix} = \mathbf{D}_1 \begin{pmatrix} i_1 \\ i_2 \end{pmatrix} \quad and \quad \begin{pmatrix} r_1 \\ r_2 \end{pmatrix} = \mathbf{D}_2 \begin{pmatrix} i_1 \\ i_2 \end{pmatrix} \tag{2.10}$$

The eigenvectors of the 2 x 2 matrix $\mathbf{D}_1$ and $\mathbf{D}_2$ represent the forward and the backward propagating eigenmodes respectively, in the isotropic medium. These eigenwaves in the isotropic medium bounding the cholesteric gives the eigenmodes which are responsible for building the standing and propagating waves inside the cholesteric medium. For example at the normal incidence the eigenvectors of the $\mathbf{D}_1$ and $\mathbf{D}_2$ matrix are circular waves. As mentioned earlier the superposition of these waves inside the cholesteric medium gives rise to a linearly polarized standing wave.

2.3 Anomalous transmission in first order Bragg reflection

To look for the anomalous transmission at the oblique incidence it is necessary to first know the eigenmodes inside the mediurn for different angles of incidence (θ_i) of the incident light. Our calculations show that at small angles θ_i < 30" the eigenmodes deviates little from the circular state. At the intermediate angles (30° *to* 50°) the eigenvectors of $\mathbf{D_1}$ and $\mathbf{D_2}$ change even within the reflection band (change with wavelength) and are in general elliptical. For large angles of incidence the eigenvectors of $\mathbf{D_1}$ and $\mathbf{D_2}$ matrices are predominantly linear, and the nature of the eigenmodes does not change within the reflection band. This is in accordance with the calculations of Miraldi et.al. [12]. Also at these large angles the Bragg band splits into three sub-bands. This is shown in the dispersion curve computed for the incidence angle θ_i = 60" in figure 2.2. In the first and third sub-band we have a pair of attenuated and propagating eigenmodes. The middle sub-band is a non-Bragg band. Here all the eigenmodes are attenuated. In this sub-band an incident polarization always get reflected to its orthogonal polarization. The reflectance of the TE and TM polarization in the non-absorbing cholesteric is shown in figure **2.3.** Though the TM wave is highly reflected in the short wavelength sub-band, it gets transmitted in the long wavelength sub-band. On the other hand it is TE that is highly reflected in the long wavelength sub-band.

In view of these polarization features we conclude that the TE and TM waves should he used to study the anomalous transmission in the absorbing cholesteric

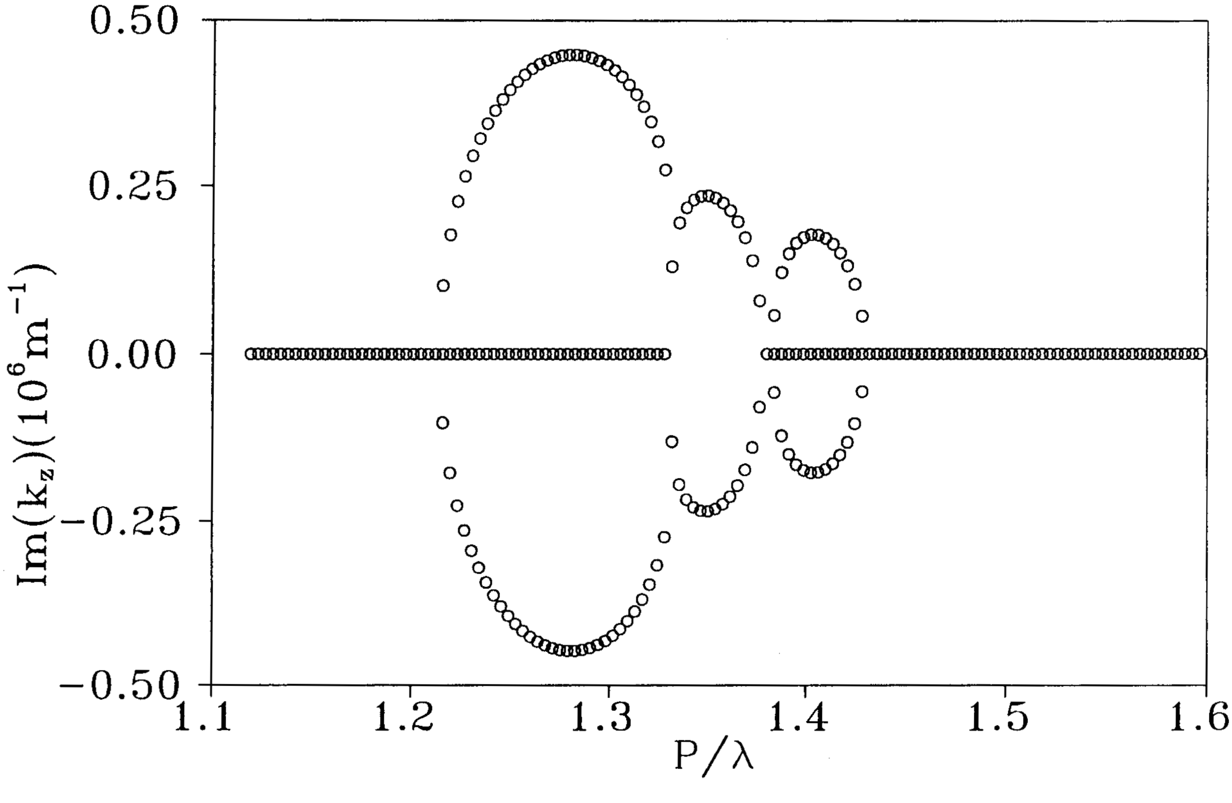

Figure 2.2: The imaginary part of block wavevectors plotted as a function of P/λ at $\theta_i = 60°$ for the first Bragg band in a non-absorbing cholesteric.

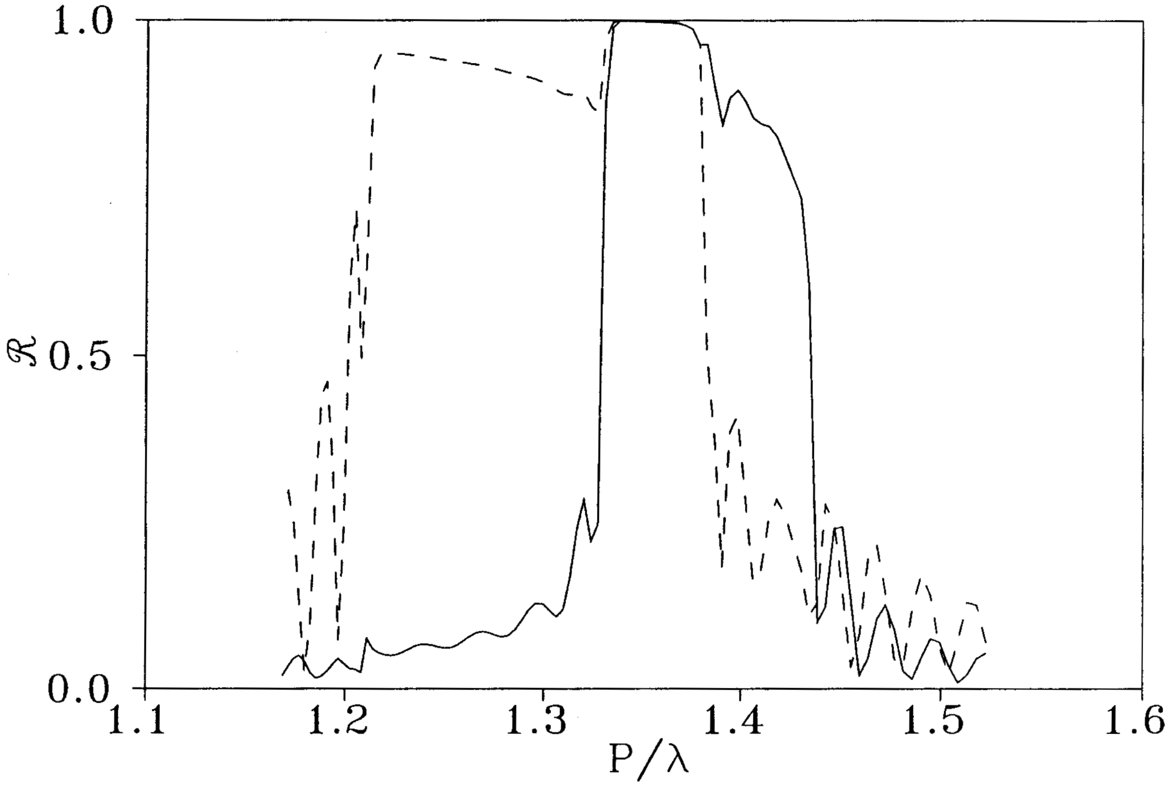

Figure **2.3:** Reflectance of TM (solid curve) and TE (long dashed curve) waves for a non-absorbing cholesteric at $\theta_i = 60°$ in the first Bragg band.

media at large angles. If we consider positive linear dichroism in the cholesteric medium then the propagation of the TE and TM waves through this medium will be such that in the short wavelength sub-band the TM wave which is more reflected also shows an enhanced transmission. This is shown in figure 2.4a and 2.4b. On the other hand if the linear dichroism is negative then our calculation show that the TE wave gets anomalously transmitted in the long wavelength sub-band.

The anomalous transmission has been experimentally well studied for normal incidence [5]. But at oblique incidence, there are not many experimental investigations to observe this effect. One attempt was made by Endo et.al. [13]. In their experiments they used RC and LC waves and found that the anomalous transmission occurs only at small angles ($0°$ *to* $19°$) and not at higher angles. We can understand their experimental results in the light of our theoretical results. Since the nature of the eigenwaves deviates little from the circular nature at small θ_i, it was possible for Endo et. al. to observe the anomalous transmission at small angles. But for high θ_i, the circular waves are no longer the eigenmodes. Hence studying tlie effect at high angles of incidence with circular waves is not appropriate. One has to choose the polarization of the incident wave to be the same as that of the eigenrriodes inside the medium, to observe the anomalous transmission for high angles of incidence. This is the main reason for Endo et.al. not observing the effect experimentally at high angles of incidence. Here we have shown theoretically that the anomalous transmission does exist at high angles but for the linearly polarized light.

Some of the optical properties of normal cholesterics at oblique incidence are very similar to those of the field induced cholesteric soliton structure [14,15]. For

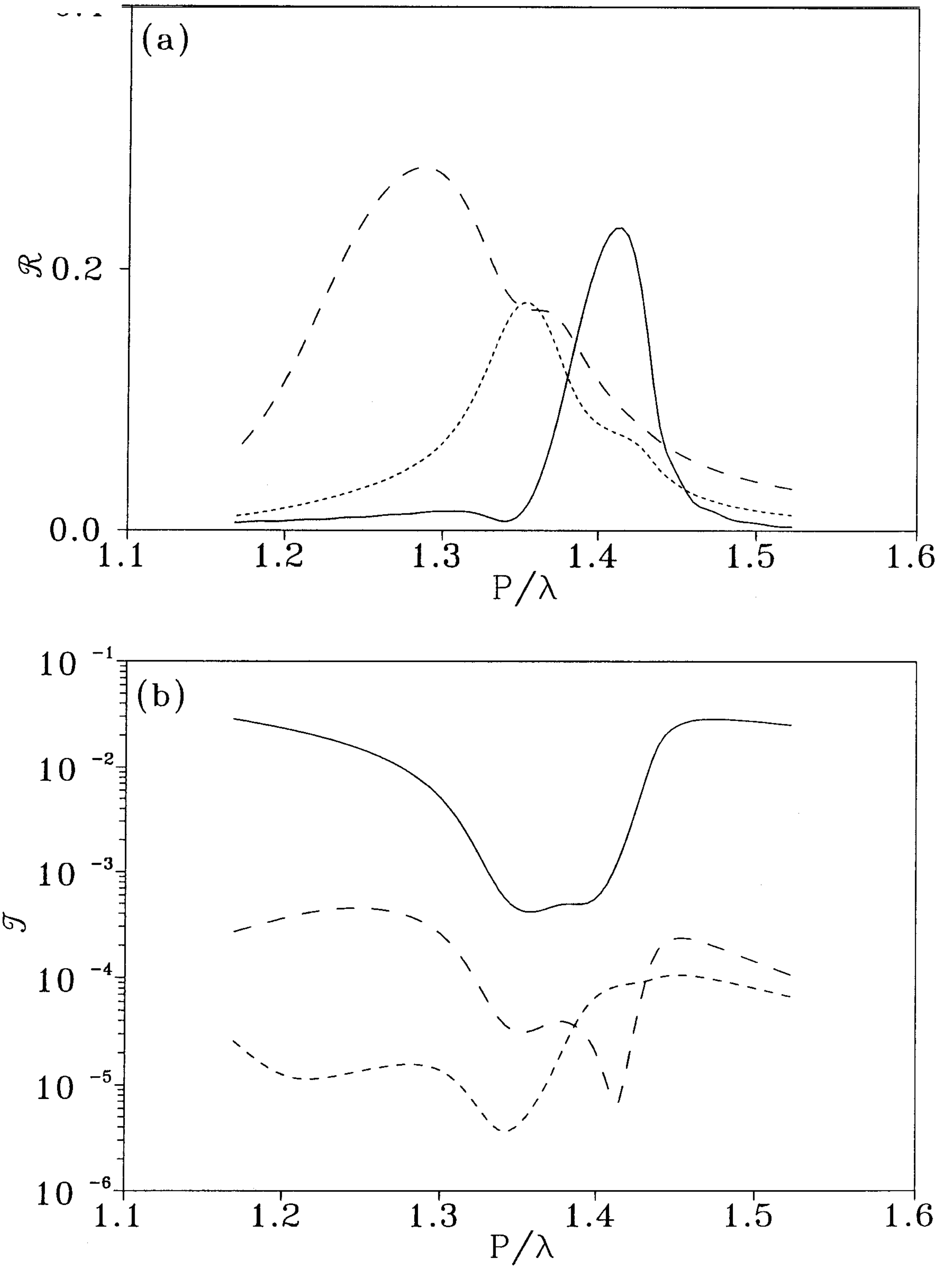

Figure 2.4: (a)Reflectance of TM-TM (solid curve), TE-TM (short dashed curve), TE-TE (long dashed curve) waves (b) Transmittance of TM-TM ,TE-TM and TE-TE waves for an absorbing cholesteric at $\theta_i = 60°$ in the first Bragg band.

example, higher order Bragg reflections, splitting of Bragg bands and anomalous transmission are some of the features common in both the cases.

For the normal incidence of light, LC wave does not sense the twisted structure of the cholesteric medium and experiences only average absorption. Hence the transmission of the RC wave can be compared to the LC wave and it is the analogue of the Borrmann effect in X-rays [2]. At high angles both the eigenwaves TE and TM, sense the cholesteric structure, and therefore the transmitted intensity of the two waves can not be compared to bring in an analogy with Borrmann effect. Therefore we take the value of the mean absorption coefficient to be equal to $[Im(\sqrt{\epsilon_1}) + Im(\sqrt{\epsilon_2})]/2$ which happens to be the absorption coefficient for the LC wave at normal incidence. Using this value as reference, we calculate the transmittance at oblique incidence for the TM wave through an isotropic medium. This is analogous to the transmittance of the X-ray through an absorbing crystal not at Bragg setting. In figure 2.5 we compare this transmittance with the transmittance of the TM wave through the cholesteric medium and bring out the analogy with the Borrmann effect. We find that the TM wave is anomalously transmitted over the entire Bragg band exhibiting Borrmann effect. In the case of molecules having negative linear dichroism, a similar effect can be noticed for the TE wave.

2.4 Anomalous transmission in higher order Bragg reflections

Like the first order Bragg reflection, the second order also has three sub-bands. In the presence of positive linear dichroism the TM wave experiences less attenuation throughout the reflection band and is transmitted more than the TE wave. In fact,

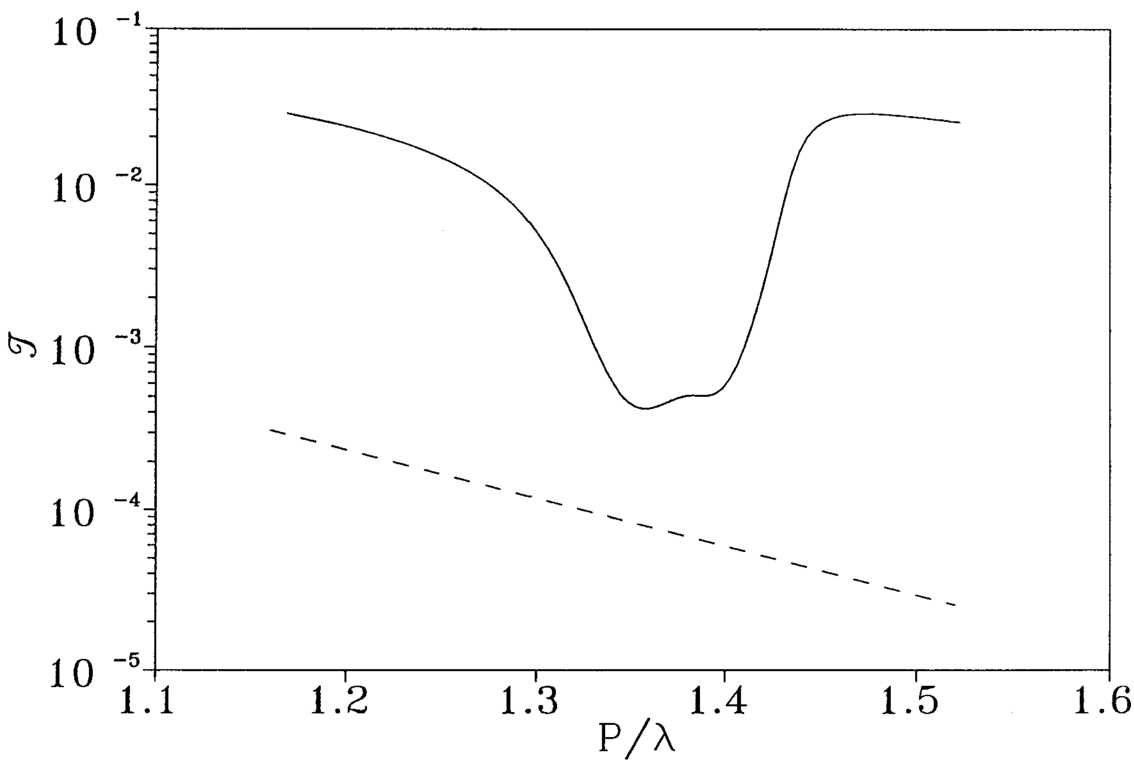

Figure 2.5: The comparison of the transmittance of the TM wave (solid curve) through the absorbing cholesteric medium with that of the same wave through an isotropic medium (dashed curve) with same absorption as that of the cholesteric medium at $\theta_i = 60°$.

the transmittance of the TE wave is almost zero (see figure 2.6a). For comparison the reflectance for the TE and TM waves is shown in figure 2.6b. In this order, the anomalous transmission for the TM wave in short wavelength sub-band is not as pronounced as in the first order. Similarly with negative linear dichroism, we find the TE wave getting anomalously transmitted in the long wavelength sub-band as in the first order reflection. We can expect anomalous transmission in higher orders but the effect will be less pronounced in successive higher orders.

2.5 Effect of absorption on the reflectance of the non-Bragg band

In the non-absorbing cholesterics at oblique incidence we find the splitting of the reflection band into three sub-bands. In the central reflection region, called the non-Bragg reflection band, the reflectance is always unity and is independent of the polarization of the incident light. Our theory leads to an interesting result. We find that at oblique incidence in the presence of linear dichroism, the reflectivity in this central sub-band to be very much dependent on the state of polarization of the incident light. This dependence of the reflectance on the polarization can be well understood by looking at the intensity of the electric field of the non-propagating eigenmodes. In the absence of absorption the fields of both the non-propagating eigenmodes suffer attenuation to the same extent as shown in figure 2.7. But in the presence of absorption the two eigenmodes see the absorption of the medium differently and one eigenmode gets attenuated more than the other. Hence the reflectance becomes polarization dependent.

In conclusion our studies show that anomalous transmission at oblique incidence

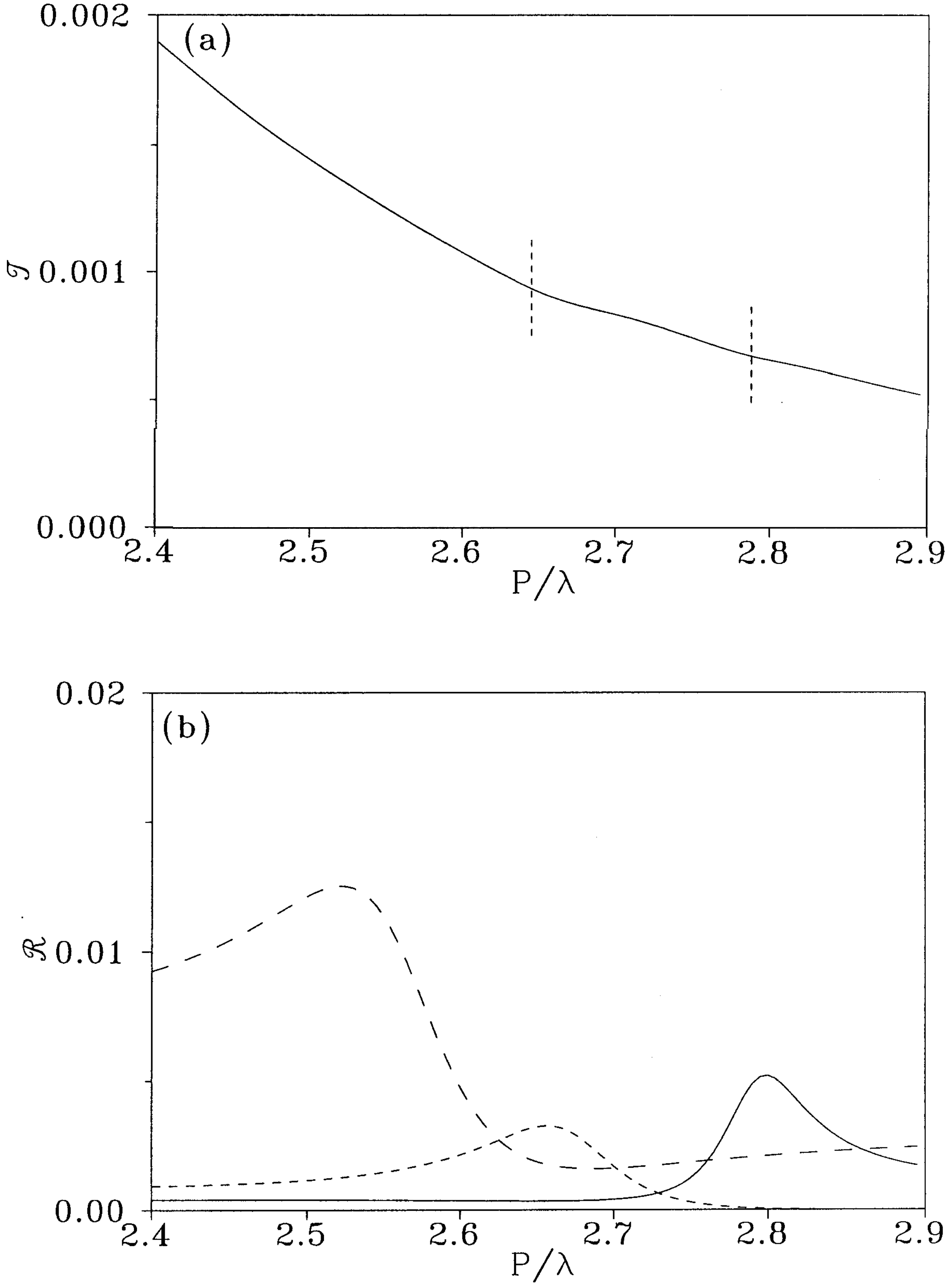

Figure 2.6: (a) Transmittance of TM (solid curve) wave. The dashed vertical lines indicate the region of anomalous transmission. Here the transmittance for the TE wave is almost zero. (b) Reflectance of TM-TM (solid curve), TE-TM (short clashed curve), TE-TE (long dashed curve) waves for an absorbing cholesteric at $\theta_i = 60^o$ in the second order Bragg reflection.

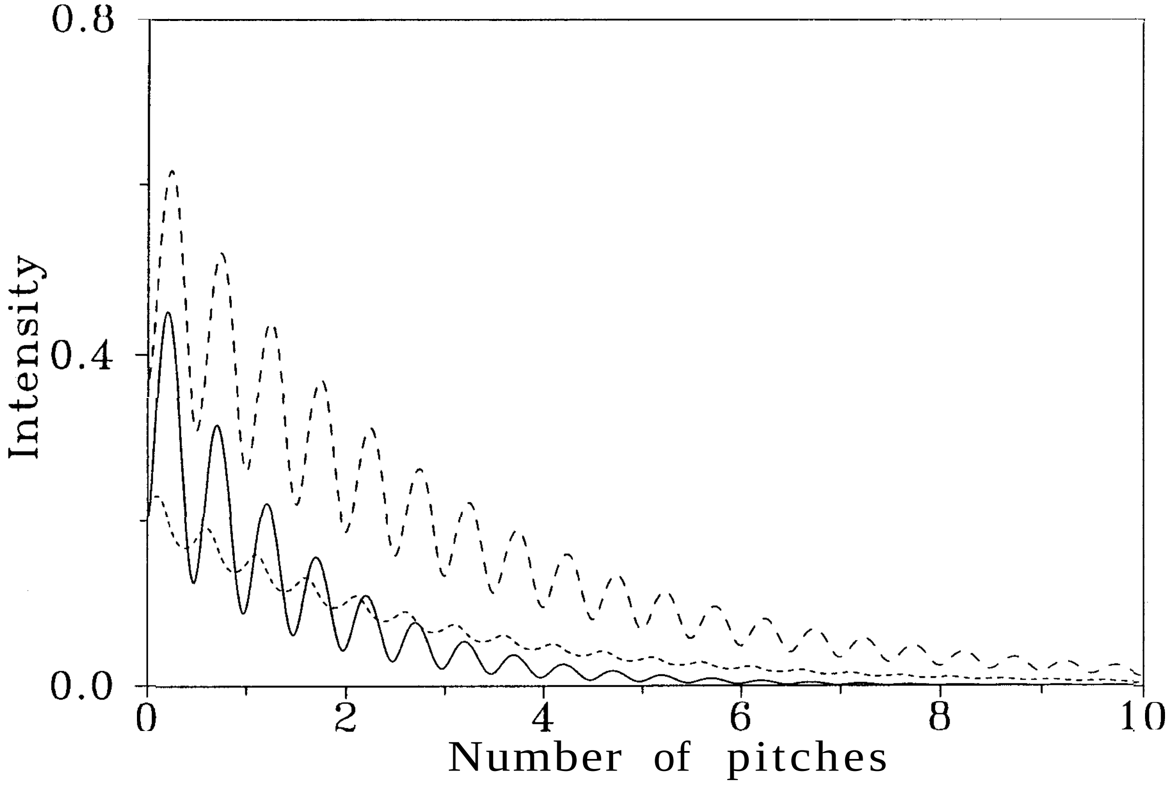

Figure 2.7: Intensity of the E field of the two nonpropagating modes in the non-Bragg region for non-absorbing (long-dashed curve) cholesteric as a function of number of pitches (in this case the two modes are attenuated by the same amount) and the intensity of the E field for the same two modes (solid and short-dashed curves) in the same region for the absorbing cholesteric medium at $\theta_i = 60°$.

is very sensitive to the polarization of the incident wave. We also find that at high angles, ***TM*** and TE waves suffers anomalous transmission in the short and long wavelength sub-band respectively depending on the sign of linear dichroism. A similar effect is also be found in higher order Bragg bands. In the absorbing case the reflectance of the non Bragg band becomes dependent on the polarization of the incident light.

References

[1] E. Sackmann and L. Voss," Circular *dichroism* of helically arranged *molecules* in cholesteric phases", Chem. Phys. Lett., 14, 528 (1972).

[2] R.W.James, The optical Principles of the Diffraction of X-rays, G.Bell and Sons Ltd, London, 1967.

[3] R.Nityananda, U.D.Kini, S.Chandrasekhar, K. **A.** Suresh "Anomalous transmission (Borrmann effect) in absorbing cholesteric liquid crystals", Proceedings of the International Liquid Crystal Conference, Bangalore, December 3-8, 1973, *Pramana* Suppl., **1**, 325 (1975).

[4] S.Chandrasekhar, G.S.Ranganath and K. **A.** Suresh " Dynamical theory *of* reflection from cholesteric liquid crystals", Proceedings of the International Liquid Crystal Conference, Bangalore, December 3-8, 1973, Pramana Suppl., **1,** 341 (1975).

[5] K.**A**.Suresh, "*An experimental* study of anomalous *transmission (Borrmann* effect) in absorbing *cholesteric* liquid crystals", Mol. Cryst. Liquid Cryst., 35, 267 (1976).

[6] R.Nityananda, "On the theory of light propagation in cholesteric liquid crystals", Mol. *Cryst.* Liquid *Cryst.*, 21, 315 (1973).

[7] R.Dreher and G.Meier, "Optical properties of cholesteric liquid crystals ", Phys. Rev A, **8,** 1616 (1973).

[8] C.Oldano, E.Miraldi and P.T.Valabrega "Dispersion relation for propagation of light in cholesteric liquid crystal", Phys. *Rev.A*, 27, 3291 (1983).

[9] C.Oldano, "Many-wave approximation for light propagation in cholesteric liquid crystals", Phys. *Rev.A*, 31, 1014 (1985).

[10] D.W.Berreman, "Optics in stratified and anisotropic medium: 4×4- matrix formulation", J. Opt. *Soc.* Ame., 62, 502 (1972).

[11] C.Oldano, "Electromagnetic wave propagation in anisotropic stratified media", Phys. *Rev.A*, 40, 6014 (1989).

[12] E.Miraldi, C.Oldano, P.I.Taverona and L. Trossi, "Optical properties of cholesteric liquid crystal at oblique incidence", Mol. *Cryst.* Liquid *Cryst.*, 103, 155 (1983).

[13] S.Endo, T.Kuribara and T.Akahane, "**A** study of the anomalous transmission (Borrmann effect) on obliquely incident light in an absorbing single-domain cholesteric liquid crystal", Jpn. **J.** Appl. Phys., 22, L499 (1983).

[14] P.B. Sunil Kumar and G.S.Ranganath, "Structural and optical behavior of *cholesteric* soliton lattices", J. *Phys. II* France, 3, 1497 (1993)

[15] S. Shtrikman and M. Tur, "*Optical properties* **of** *the distorted cholesteric structure* ", **J. Opt.** *Soc. Ame.*, **64, 1178 (1974).**