

The theory of reflexion and transmission by plane parallel cholesteric films

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Abstract. The theory of propagation in an infinite cholesteric medium is applied here to the problem of a sample bounded by plane parallel surfaces normal to the optic axis. For each circular polarization at normal incidence, the reflected and transmitted waves are found to consist of both circular polarizations. Thus, four coefficients of reflection and transmission are needed to describe the problem fully. These have been calculated in a closed analytical form, which has the correct behaviour in various limiting cases. Numerical computations are used to investigate the effect of finite sample thickness in modifying the rotation and circular dichroism predicted from the infinite medium theory. This is of importance in interpreting the results of experiments.

Introduction

The theory of the propagation of light along the axis of a cholesteric liquid crystal was first given by de Vries¹ using the Oseen² model for the dielectric tensor, subject to certain approximations. (For a discussion of these approximations, see, Nityananda³, hereafter referred to as I). Recently, an exact solution of this problem was given by Kats⁴ and by Nityananda³. We now use that solution to calculate the reflection and transmission coefficients of a plane parallel cholesteric film. These were approximately calculated by de Vries. The novel feature of our calculation is the presence of both circular polarisations in the reflected and transmitted beams, even if only one circular polarisation is incident. Thus we define 'diagonal' and 'off diagonal' coefficients r_{++} and r_{-+} where, by convention, the second suffix gives the sense of rotation of the electric vector of the incident wave, and the first the sense of the reflected (or transmitted) wave. For example, t_{+-} is the amplitude of the (+) circular component of the transmitted light for the unit incident amplitude of (-) light. The convention is the same as in I, with (+) denoting rotation in the clockwise sense in the xy -plane. This would be called a right circular wave if it travelled along $+z$, but left circular if the wave vector were along $-z$. For a transparent dielectric at normal incidence, (+) is reflected as (+) and (-) as (-). A right handed cholesteric liquid

crystal, studied at a wavelength near the reflection band, reflects (+) as predominantly (-). In his calculation of the reflection and transmission coefficients, de Vries¹ assumed that the medium on either side of the cholesteric film had a refractive index equal to the average for the cholesteric. Further, the birefringence δn was treated as a small quantity—an approximation which is certainly valid in practice. Under these conditions, the transmitted wave has the same circular polarisation as the incident wave and one circular wave is reflected, completely reversed in sense. This agrees with experimental observations, which approach the conditions assumed since the liquid crystal is enclosed by glass slides. The work of Chandrasekhar and Prasad⁵ based on the dynamical theory led to similar results.

The emphasis in the present paper is on removing the restrictions in earlier treatments. Thus, the birefringence could be large (as is sometimes attained by dissolving optically active molecules in a nematic to form a cholesteric) and the wavelength far from, or close to the Bragg reflection. The refractive index of the surrounding medium can be arbitrary. This additional generality has not led to qualitatively new results, however. Indeed, Chandrasekhar *et al.*⁸ have shown that the dynamical theory leads to close quantitative agreement with the exact solution presented here in practical cases. The coefficients like r_{++} which were neglected in earlier treatments would describe Fresnel reflection for an isotropic dielectric, and would be of the order of the refractive index difference between the cholesteric and the glass slide. A coefficient like t_{+} would vanish for an isotropic dielectric—the formulae given below show it to be of order δn (this is the local birefringence of the cholesteric). When we come to intensities these estimates are naturally squared, so that neglecting them has not caused serious error in the earlier work.

2. Calculation of the reflection and transmission coefficients

The sample, of thickness T , is assumed to occupy the region between the planes $z = 0$ and $z = T$, with its helical axis parallel to z , *i.e.*, normal to the plane of the sample. This is called the plane texture and is a commonly used experimental geometry. The incident wave is taken to be a general superposition of (+) and (-) circular polarisations, with complex amplitudes E_{+i} and E_{-i} . (We use the suffix 'i' for incident, 'r' for reflected, and 't' for transmitted light.) Its wavevector is $(0, 0, K)$ where $K = \omega/c$ if the region $z < 0$ is free space.

The reflected wave has wave vector $(0, 0, -K)$ and is a superposition of both circular components with amplitudes E_{+r} and E_{-r} . In the region $z > T$, we have a transmitted wave with wave vector $(0, 0, K)$ and circular components $E_{\pm t}$. In what follows we use the notation of I in which the two circular components are written together in the form of a

row or column vector (1×2 or 2×1 matrix). Thus, the incident wave is written as

$$[E_{+1} e^{iK_1 z}, E_{-1} e^{iK_1 z}].$$

The reflected and transmitted waves are written similarly (figure 1).

We now have to match these waves outside the cholesteric to a suitable solution of Maxwell's equations within. This solution is given in the work of Kats⁴ and in that of Nityananda³. From general considerations we see that there must be four independent solutions for propagation along the axis because we have two coupled second order differential equations (essentially the wave equation $\nabla \times (\nabla \times \mathbf{E}) = -\frac{1}{c^2} \hat{\epsilon} \frac{\partial^2 \mathbf{E}}{\partial t^2}$ in component form) for E_x and E_y or E_+ and E_- . For example, in an isotropic dielectric, these four solutions could just be two linearly polarised waves along $+z$ and two more along $-z$. For the cholesteric, the four independent solutions are

$$U_{t+} = [e^{iK_1 z}, d e^{i(K_1 - 2q) z}]$$

$$U_{b+} = [d e^{-i(K_1 - 2q) z}, e^{-iK_1 z}]$$

$$U_{t-} = [f e^{i(K_1 + 2q) z}, e^{iK_1 z}]$$

$$U_{b-} = [e^{-iK_1 z}, f e^{-i(K_1 + 2q) z}]$$

We choose a linear combination of these four solutions with coefficients $t_{\pm}, t_{b\pm}$ to describe the electric field in the medium*. We have four boundary conditions to fulfil at each interface; one for each cartesian or circular component of the electric and magnetic fields. The magnetic field can be expressed in terms of the electric field using Maxwell's equations. Because the fields have no dependence on x and y , and no z components, these reduce to

$$\frac{i\omega}{c} B_x = i(-k_z E_y)$$

$$\frac{i\omega}{c} B_y = i(k_z E_x)$$

In terms of the circular components $E_{\pm} = \frac{E_x \pm iE_y}{\sqrt{2}}$ we get $(\omega/c) B_{\pm} = \pm i k_z E_{\pm}$. The factor of $\pm i$ means that the magnetic field is at right

* The expressions for K_1 , K_2 , d and f are given in Appendix I along with the reason for the notation.

angles to the rotating electric field and hence leads or lags it by 90° in phase. Apart from a constant factor, we get the circular components of the magnetic field by multiplying those of the electric field by the wave vector. We match these on the two sides of each interface, along with the electric fields themselves.

$$\begin{array}{c}
 \begin{array}{c} \rightarrow \\ \left[\begin{array}{c} E_{+i} \\ E_{-i} \end{array} \right] \exp(iKz) \\ \leftarrow \\ \left[\begin{array}{c} E_{+r} \\ E_{-r} \end{array} \right] \exp(-iKz) \end{array} \quad \left| \begin{array}{c} t_{f+} \left[\begin{array}{c} \exp(iK_1 z) \\ d \exp[i(K_1 - 2q)z] \end{array} \right] \\ t_{f-} \left[\begin{array}{c} f \exp[i(K_2 + 2q)z] \\ \exp(iK_2 z) \end{array} \right] \end{array} \right. + \\
 \text{Semi-infinite Sample}
 \end{array}$$

$$\begin{array}{c}
 \begin{array}{c} \rightarrow \\ \left[\begin{array}{c} E_{+i} \\ E_{-i} \end{array} \right] \exp(iKz) \\ \leftarrow \\ \left[\begin{array}{c} E_{+r} \\ E_{-r} \end{array} \right] \exp(-iKz) \end{array} \quad \left| \begin{array}{c} t_{f+} \left[\begin{array}{c} \exp(iK_1 z) \\ d \exp[i(K_1 - 2q)z] \end{array} \right] + t_{f-} \left[\begin{array}{c} f \exp[i(K_2 + 2q)z] \\ \exp(iK_2 z) \end{array} \right] \\ t_{b+} \left[\begin{array}{c} \exp(-iK_2 z) \\ f \exp[-i(K_2 + 2q)z] \end{array} \right] + t_{b-} \left[\begin{array}{c} d \exp[i(K_1 - 2q)z] \\ \exp(-iK_1 z) \end{array} \right] \end{array} \right. \rightarrow \left[\begin{array}{c} E_{+t} \\ E_{-t} \end{array} \right] \exp(iKz) \\
 \text{Finite Sample}
 \end{array}$$

Figure 1 The solution of Maxwell's equations for a semi-infinite and finite cholesteric film at normal incidence. The coefficients $t_{f\pm}$, $t_{b\pm}$, $E_{\pm r}$ and $E_{\pm t}$ have to be determined from the boundary conditions.

The solutions in the three regions are given in figure 1. The matching conditions, which are written down in table 1 constitute eight equations for the eight unknown quantities in our solution $E_{\pm r}$, $E_{\pm t}$, $t_{f\pm}$ and $t_{b\pm}$. The last four appear as intermediate variables only, and the final aim is to express $E_{\pm r}$ and $E_{\pm t}$ in terms of $E_{\pm i}$, which are assumed to be known. The coefficients in these expressions are identified with the quantities r_{++} etc. defined in the introduction. Thus,

$$E_{+r} = r_{++} E_{+i} + r_{+-} E_{-i}$$

$$E_{-r} = r_{-+} E_{+i} + r_{--} E_{-i}$$

A similar pair of equations relates $E_{\pm t}$ to $E_{\pm i}$, viz.,

$$E_{+t} = t_{++} E_{+i} + t_{+-} E_{-i}$$

$$E_{-t} = t_{-+} E_{+i} + t_{--} E_{-i}.$$

Table 1. Boundary conditions for the electric field

At $z = 0$:

$$E_{+i} + E_{+r} = t_{i+} + ft_{i-} + t_{b+} + dt_{b-} \quad (1)$$

$$E_{-i} + E_{-r} = dt_{i+} + t_{i-} + ft_{b+} + t_{b-} \quad (2)$$

At $z = T$:

$$\begin{aligned} E_{+i} \exp(iKT) &= t_{i+} \exp(iK_1T) + ft_{i-} \exp[i(K_2 + 2q)T] \\ &+ t_{b+} \exp(-iK_2T) + dt_{b-} \exp[-i(K_1 - 2q)T] \end{aligned} \quad (3)$$

$$\begin{aligned} E_{-i} \exp(iKT) &= dt_{i+} \exp[i(K_1 - 2q)T] + t_{i-} \exp(iK_2T) \\ &+ ft_{b+} \exp[-i(K_2 + 2q)T] + t_{b-} \exp(-iK_1T) \end{aligned} \quad (4)$$

Boundary conditions for the magnetic field

At $z = 0$:

$$K(E_{+i} - E_{+r}) = K_1t_{i+} + f(K_2 + q)t_{i-} - K_2t_{b+} - (K_1 - 2q)dt_{b-} \quad (1')$$

$$K(E_{-i} - E_{-r}) = d(K_1 - 2q)t_{i+} + K_2t_{i-} - f(K_2 + 2q)t_{b+} - K_1t_{b-} \quad (2')$$

At $z = T$:

$$\begin{aligned} KE_{+i} \exp(iKT) &= K_1t_{i+} \exp(iK_1T) + f(K_2 + 2q)t_{i-} \exp[i(K_2 + 2q)T] \\ &- K_2t_{b+} \exp(-iK_2T) - (K_1 - 2q)dt_{b-} \exp[-i(K_1 - 2q)T] \end{aligned} \quad (3')$$

$$\begin{aligned} KE_{-i} \exp(iKT) &= d(K_1 - 2q)t_{i+} \exp[i(K_1 - 2q)T] + K_2t_{i-} \exp(iK_2T) \\ &- f(K_2 + 2q)t_{b+} \exp[-i(K_2 + 2q)T] - K_1t_{b-} \exp(-iK_1T) \end{aligned} \quad (4')$$

Eliminating $t_{i\pm}$ and $t_{b\pm}$ as indicated, we obtain expressions for the reflexion and transmission coefficients, which are given in table 2. It is convenient to define various intermediate quantities A , B , X and Y with suffixes from 1 to 4, D and Δ . This is solely to shorten the expressions for r and t . The next section gives the results of numerical calculations based on these formulae.

Table 2

$$A_1 = \frac{1}{2} \left(1 + \frac{K_1}{K} \right), A_2 = \frac{f}{2} \left(1 + \frac{K_2 + 2q}{K} \right), A_3 = \frac{1}{2} \left(1 - \frac{K_2}{K} \right),$$

$$A_4 = \frac{d}{2} \left[1 - \frac{(K_1 - 2q)}{K} \right]$$

$$B_1 = \frac{1}{2} \left(1 - \frac{K_1}{K} \right), B_2 = \frac{f}{2} \left[- \frac{K_2 + 2q}{K} \right], B_3 = \frac{1}{2} \left[1 + \frac{K_2}{K} \right],$$

$$B_4 = \frac{d}{2} \left[1 + \frac{(K_1 - 2q)}{K} \right]$$

$$X_1 = (K_1^2 - K^2) - d^2 (K_1 - 2q + K) (K_1 - 2q - K)$$

$$X_2 = [f(K + K_1) (K_2 + 2q - K) - d(K + K_1 - 2q) (K_2 - K)] \\ \exp[-i(K_1 - 2q - K_2)T]$$

$$X_3 = [d(K_1 - 2q - K) (K + K_2) - f(K + K_2 + 2q) (K_1 - K)] \\ \exp[i(K_1 - 2q - K_2)T]$$

$$X_4 = (K_2^2 - K^2) - f^2 (K_2 + 2q + K) (K_2 + 2q - K)$$

$$D = \exp[-i(K_1 + K_2)T] [(K + K_1) (K + K_2) \\ - df(K + K_2 + 2q) (K + K_1 - 2q)]$$

$$\Delta = (A_1 D + A_3 X_1 + A_4 X_3) (B_3 D + B_2 X_2 + B_1 X_4) \\ - (A_2 D + A_3 X_2 + A_4 X_4) (B_4 D + B_2 X_1 + B_1 X_3)$$

$$Y_1 = D(B_3 D + B_2 X_2 + B_1 X_4), Y_2 = -D(A_2 D + A_3 X_2 + A_4 X_4)$$

$$Y_3 = -D(A_2 D + A_3 X_2 + A_4 X_4), Y_4 = D(A_1 D + A_3 X_1 + A_4 X_3)$$

$$r_{++} = \frac{B_1 Y_1 + B_2 Y_3}{\Delta} + \frac{1}{D\Delta} [B_3 (X_1 Y_1 + X_2 Y_3) + B_4 (X_3 Y_1 + X_4 Y_3)]$$

$$r_{+-} = \frac{B_1 Y_2 + B_2 Y_4}{\Delta} + \frac{1}{D\Delta} [B_3 (X_1 Y_2 + X_2 Y_4) + B_4 (X_3 Y_2 + X_4 Y_4)]$$

$$r_{-+} = \frac{A_4 Y_1 + A_3 Y_3}{\Delta} + \frac{1}{D\Delta} [A_2 (X_1 Y_1 + X_2 Y_3) + A_1 (X_3 Y_1 + X_4 Y_3)]$$

$$r_{--} = \frac{A_4 Y_2 + A_3 Y_4}{\Delta} + \frac{1}{D\Delta} [A_2 (X_1 Y_2 + X_2 Y_4) + A_1 (X_3 Y_2 + X_4 Y_4)]$$

$$t_{++} = \frac{Y_1}{\Delta} \exp [i (K_1 - K) T] + \frac{Y_3}{\Delta} f \exp [i (K_2 + 2q - K) T]$$

$$+ \frac{1}{D\Delta} [(X_1 Y_1 + X_2 Y_3) \exp \{-i (K_2 + K) T\} \\ + d (X_3 Y_1 + X_4 Y_3) \exp \{-i (K_1 - 2q + K) T\}]$$

$$t_{+-} = \frac{[Y_2 \exp \{i (K_1 - K) T\} + Y_4 f \exp \{i (K_2 + 2q - K) T\}]}{\Delta}$$

$$+ \frac{1}{D\Delta} [(X_1 Y_2 + X_2 Y_4) \exp \{-i (K_2 + K) T\} + d (X_3 Y_2 + X_4 Y_4) \\ \exp \{-i (K_1 - 2q + K) T\}]$$

$$t_{-+} = \frac{1}{\Delta} [dY_1 \exp \{i (K_1 - 2q - K) T\} + Y_3 \exp \{i (K_2 - K) T\}]$$

$$+ \frac{1}{D\Delta} [f (X_1 Y_1 + X_2 Y_3) \exp \{-i (K_2 + 2q + K) T\} \\ + (X_3 Y_1 + X_4 Y_4) \exp \{-i (K_1 + K) T\}]$$

$$t_{--} = \frac{1}{\Delta} [dY_2 \exp \{i (K_1 - 2q - K) T\} + Y_4 \exp \{i (K_2 - K) T\}]$$

$$+ \frac{1}{D\Delta} [f (X_1 Y_2 + X_2 Y_4) \exp \{-i (K_2 + 2q + K) T\} \\ + (X_3 Y_2 + X_4 Y_4) \exp \{-i (K_1 + K) T\}]$$

The experimental quantities usually studied are optical rotation and circular dichroism. The latter is usually defined by

$$D = \frac{\sqrt{I_+} - \sqrt{I_-}}{\sqrt{I_+} + \sqrt{I_-}}$$

where I_+ and I_- are the intensities of the transmitted beam for equal incident intensities of the two circular polarisations. The above definition contains the implicit provision that (+) light is transmitted

only as $(+)$ and $(-)$ light as $(-)$. Correspondingly, the experimenter does not usually analyse the transmitted light into its circular components before making intensity measurements. We have seen in the previous section that the transmitted light contains both circular polarisations. To conform to the experimental situation, we take the total intensity of these two components as I_+ or I_- in plotting the theoretical curves. Likewise, when computing optical rotation, bearing in mind that the transmitted light is in general elliptic, the major axis of the ellipse is chosen as the direction of the emergent polarisation. In all the numerical examples, the pitch P has been set equal to 0.3571μ and the birefringence $\delta n = 0.07$, the mean refractive index is 1.435 , so that the two principal refractive indices are 1.4 and 1.47 . The thickness T is variable and is indicated in each figure. The reflection band is centered at $\lambda = \bar{n}P = 0.5125 \mu$ and has a width $P\delta n \sim 0.05 \mu$.

Figure 2 shows the reflection coefficient $|r_-|^2$. This measures reflection with change in sense of the circular polarisation, which is the characteristic property of the cholesteric liquid crystal. This is for a sample of thickness 6μ , and shows the oscillations characteristic of finite samples studied by Chandrasekhar and Prasad⁵ and by Dreher, Meier and Saupe⁶. The spacing of these oscillations agree approximately with what would be calculated for a plane parallel film of isotropic dielectric with the same mean refractive index and thickness, but their amplitude is of course greatly enhanced near the reflection band.

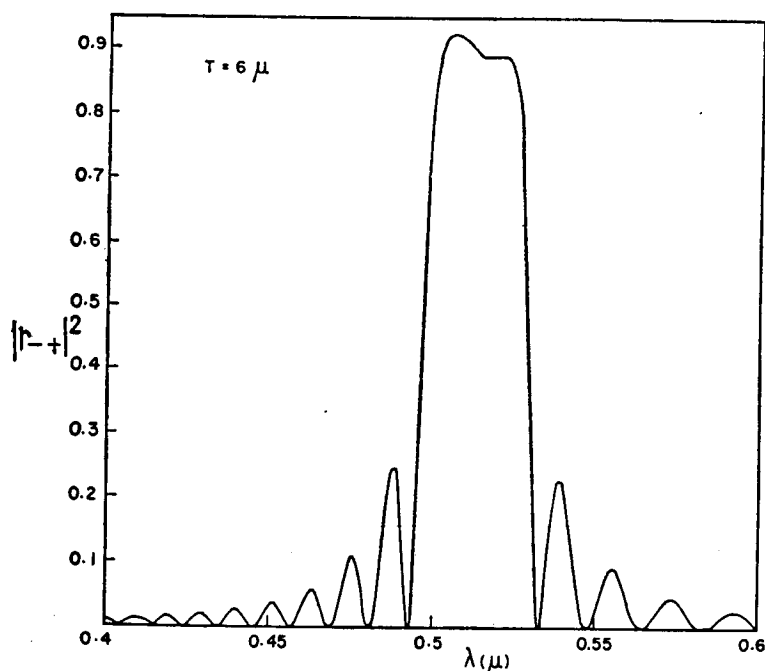


Figure 2 Intensity of reflection with change in sense of circular polarisation as a function of wavelength λ computed from the formula in table 2. The sample thickness is 6μ .

Figure 3 shows the transmitted intensity T and circular dichroism D as functions of wavelength for 4μ (figure 3a) and 6μ (figure 3b) samples. T is plotted for each circular polarisation separately, as indicated by the symbols (+) and (−) on the respective curves. As expected, the (+) circular component which is Bragg reflected shows a strong dip in transmission at the same wavelength. The circular dichroism shows a strong negative peak there, since $I_- > I_+$. The effect of increasing the sample thickness is seen by comparing figures 3a and 3b. The suppression of the R component in the transmitted beam is more complete, the negative peak in D stronger, and the oscillations more closely spaced. Further we see an approach to the case of an infinite medium which has a flat reflection maximum. The transmission curve approaches a shape with a flat minimum

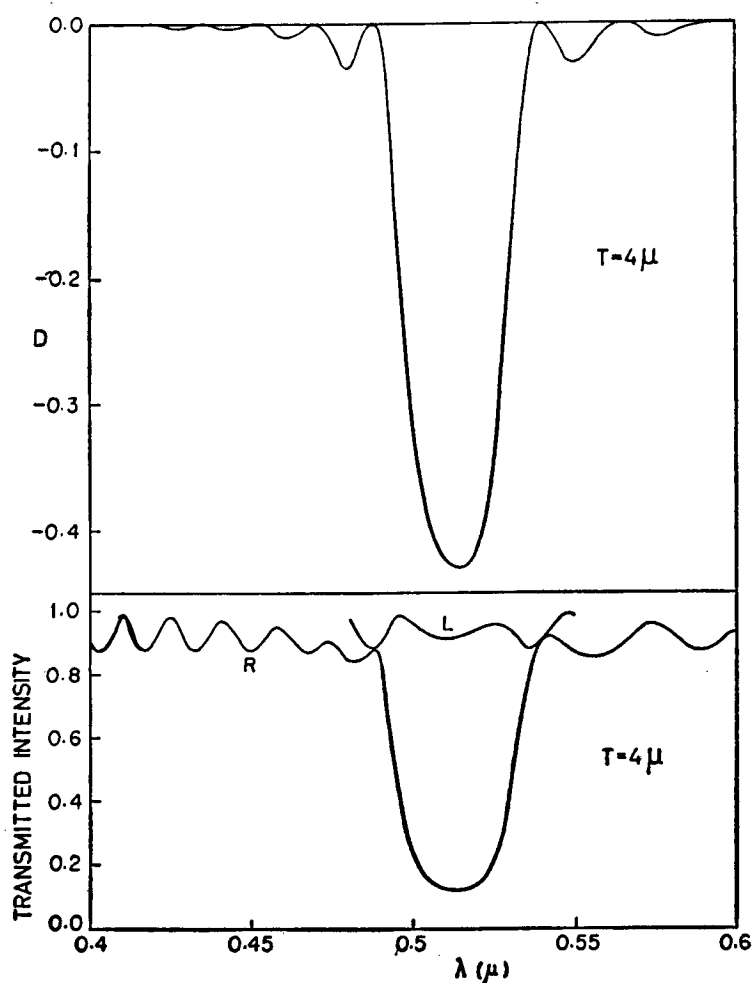


Figure 3a Transmitted intensity T and circular dichroism D computed as functions of wavelength λ . Sample thickness 4μ .

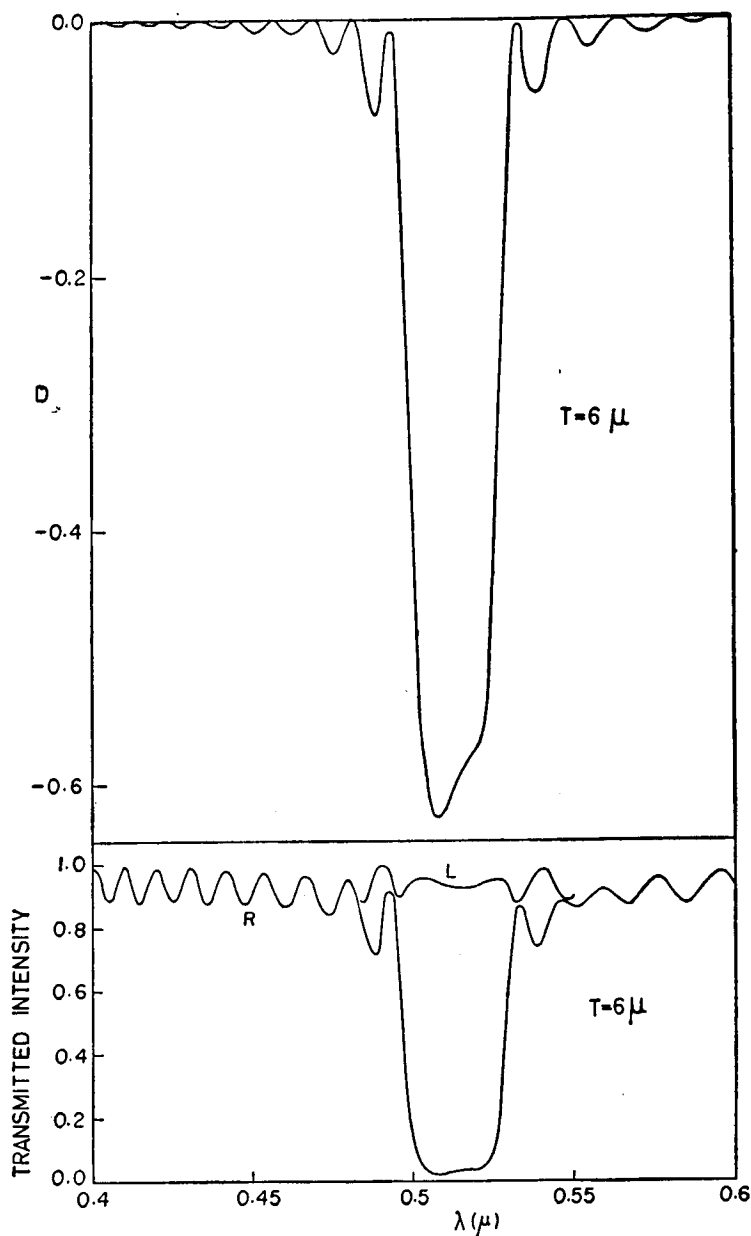


Figure 3 b Transmitted intensity T and circular dichroism D computed as functions of wavelength λ . Sample thickness 6μ .

close to zero. The extinction length of the attenuated normal wave at the centre of the reflection band is $\simeq (P/2\pi) \frac{\bar{n}}{\delta n} \sim 1.4 \mu$ in our example, increasing to ∞ at the edges of the reflection band. Therefore, the infinite medium behaviour first occurs at the centre and then spreads outwards.

It is noteworthy that the circular dichroism does not change sign away from the reflection band for a non-absorbing sample. It tends to zero as the values of T for $(+)$ and $(-)$ circular polarisations tend to equality. The behaviour of absorbing samples is different and the following paper discusses it.

Finally figures 4a and 4b show optical rotation as a function of wavelength near the reflection band for thickness $1\ \mu$ and $3\ \mu$ respectively. The dispersion qualitatively resembles that for infinite samples³. However,

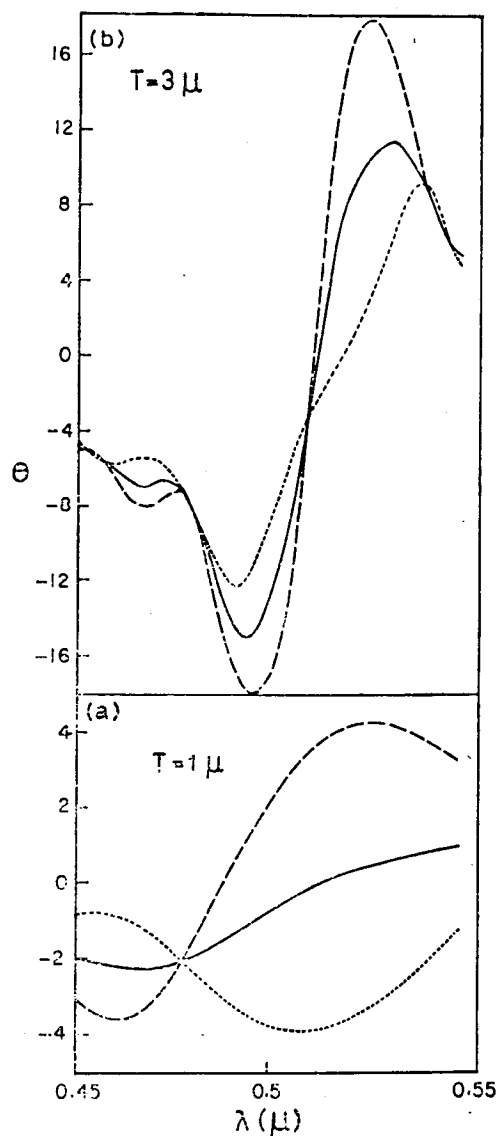


Figure 4 Optical rotation vs λ . The dotted curve is for incident linear polarisation along y , the dashed curve for x . Sample thickness (a) $1\ \mu$ (b) $3\ \mu$.

the curve for $T = 1 \mu$ (which is about 3 times the pitch) reveals that the rotation angle depends on the azimuth of the incident light. The dashed curve shows the rotation for incident linear polarisation along x and the dotted one for y . The director of the cholesteric is taken along x at the plane $z = 0$. The solid line shows the average. Even this average is not proportional to thickness—if it were the solid curves would differ only in scale. Chandrasekhar and Prasad⁵ noted the thickness dependence and studied it experimentally. Ranganath (private communication) has noted the azimuth dependence of the rotation for mixtures of right and left handed cholesterics when the pitch is much larger than the wavelength of light. This azimuth dependence is considerably reduced if the medium on either side of the cholesteric matches its average refractive index.

The behaviour of the rotatory dispersion can be correlated to that of the circular dichroism *via* the Kramers Kronig dispersion relation. As the thickness is reduced the band of circular dichroism widens and is rounded off, so that we expect the maximum and minimum of the rotation to be reduced in strength and shifted outwards. The oscillations in the circular dichroism are reflected in those of rotation*.

Summary

An exact analytical solution of the problem of reflection and transmission at normal incidence by a cholesteric film has been given. This differs from earlier treatments in including off diagonal transmission and reflection coefficients. Numerical calculations show these to be small, and confirm the results of earlier work^{1, 5, 6} on finite specimens.

Appendix I

In the infinite medium theory developed in I the normal waves were given by superpositions of two circular waves of opposite sense, with wave vectors differing by $2q$, for example

$$[e^{iK_1 z}, de^{i(K_1 - 2q)z}].$$

The term with coefficient unity is called the dominant component. There are two reasons for this—firstly, that the coefficients d and f never exceed 1 (in absolute magnitude) near the reflection band, and secondly, that they vanish in the limiting case of an isotropic dielectric ($\delta n \rightarrow 0$). In the same limiting case, K_1 reduces to $K_n = \frac{2\pi n}{\lambda}$. The suffixes b and f

* Such a correlation can only be qualitative—to our knowledge no quantitative formulation of the dispersion relation has been given for a non-uniform medium like cholesteric. While one can study the analytic properties of K_1 and K_2 , we have seen that these do not directly give rotation and circular dichroism for finite samples.

used in the text refer to 'backward' and 'forward'. The normal wave written above reduces to a circular wave propagating along $+z$ as $d \rightarrow 0$, and would hence be called 'forward'. The formulae for K_1 , K_2 , d and f are given below. ω = frequency of incident light, c = velocity of light in vacuum, ϵ_a and ϵ_b are the principal dielectric constants,

$$\epsilon = \frac{\epsilon_a + \epsilon_b}{2}, \quad \beta = \frac{\epsilon_a - \epsilon_b}{2}, \quad P = \text{Pitch}, \quad q = \frac{2\pi}{P}, \quad K = \frac{\omega}{c}, \quad K_m = \frac{\omega}{c} \sqrt{\epsilon}$$

$$K_1 = q [K_m^2 + q^2 - (4K_m^2 q^2 + \beta^2 K^4)^{1/2}]^{1/2}$$

$$K_2 = -q + [K_m^2 + q^2 + (4K_m^2 q^2 + \beta^2 K^4)^{1/2}]^{1/2}$$

$$d = \frac{K_1^2 - K_m^2}{\beta K^2}, \quad f = \frac{K_2^2 - K_m^2}{\beta K^2}.$$

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