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On the Theory of Light Propagation in Cholesteric Liquid Crystals†

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Abstract—The theory of the propagation of light along the axis of a cholesteric liquid crystal is considered. Two models have been used so far, viz., the spiralling dielectric ellipsoid model of Oseen, treated theoretically by de Vries, and the twisted pile of birefringent plates treated according to the dynamical theory by Chandrasekhar and Srinivasa Rao. The two models appear to give different results at long wavelengths, and the former seems inapplicable in the region of total reflection.

In this paper, an exact solution of the Oseen model is found, which may be used in the entire range of wavelengths, and has a simple physical interpretation. Secondly, the model of Chandrasekhar and Srinivasa Rao is shown to agree with this solution. The approximations implicit in the work of these authors and that of de Vries are clearly brought out. A numerical example illustrating the features of the exact solution is given.

1. Introduction

The optical properties of cholesteric liquid crystals have been the subject of many experimental and theoretical studies. They exhibit selective reflection of circularly polarized light of one sense within a narrow band of wavelengths $\Delta\lambda$ around a value λ_0, and a very large value of optical rotatory power which changes sign as λ crosses λ_0. These two remarkable properties are attributed to the twisted, periodic structure of the medium. A widely used model, first proposed by Oseen[1] treats the liquid crystal as an anisotropic dielectric, which can be described locally by a dielectric tensor with principal axes Oa and Ob and principal values ϵ_a and ϵ_b (Fig. 1). As we move along the z-axis (say), Oa and Ob rotate in the x-y plane

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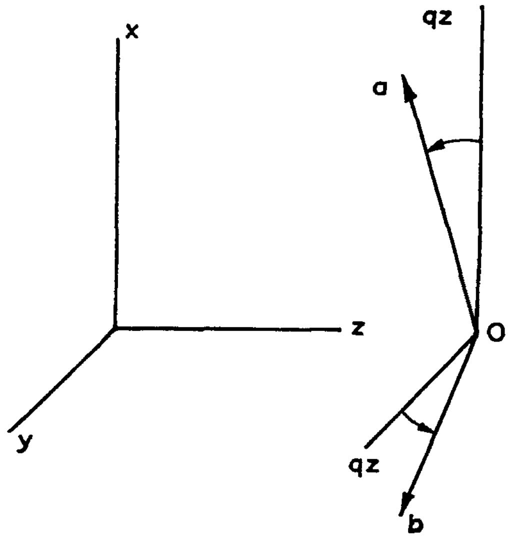

Figure 1. The Oseen model. Oa and Ob are the principal axes of the dielectric tensor at a distance z from the x-y plane.

through an angle qz. The pitch P is the distance along the z-axis corresponding to a rotation of 2π, so that $P = 2\pi/q$.

An approximate treatment of this model was given in the pioneering work of de Vries.[2] This explained the selective reflection as well as the rotation well away from λ_0. The expression derived for the rotatory power is infinite for $\lambda = \lambda_0$ and is clearly not valid near and within the reflection band.

A different approach, analogous to the dynamical theory of X-ray diffraction as formulated by Darwin,[3] was given by Chandrasekhar and Srinivasa Rao[4] (CS). The first step in their work was to calculate the modification of the state of polarization of an incident wave by a helically arranged pile of thin birefringent plates. This was carried out using the Jones calculus for optical systems, and gave rise to optical rotation—i.e., a phase difference between waves of opposite circular polarization, agreeing with the earlier results of Mauguin.[5] The second stage was to account for the effect of multiple reflection of the waves at the interfaces between the plates. An approximate calculation showed that one turn of the helix reflects the circular wave whose sense is opposite to that of the structure with a reflection coefficient iQ and transmits the other wave entirely.

This reflection coefficient was used in the difference equation connecting the amplitudes of the wave at the $(n-1)^{\text{th}}$, n^{th} and $(n+1)^{\text{th}}$ layers, each layer introducing a phase difference between the two waves as calculated earlier.

CS could explain the presence of a reflection band as well as the anomalous rotatory dispersion and circular dichroism within this band. Their expression for the rotatory power reduced that of Mauguin for $\lambda \ll \lambda_0$. However it predicted a second change of sign at $\lambda > \lambda_0$ in contrast to the de Vries theory.

Chandrasekhar and Shashidhara Prasad[6] undertook a search for this reversal of the rotation in the infrared region of the spectrum, but their experiments showed that the rotation approached zero with increasing wavelength without changing sign. Further, they refined the theory by taking into account (i) the wavelength dependence of Q, (ii) the finite size of the crystal, and concluded that, under these conditions, the change in sign might occur only at very long wavelengths. They recognized, however, that the discrepancy with de Vries theory remained in principle.

The later work of Aihara and Inaba[7] is based on a computational approach. Recently Marathay[8] has given an exact treatment of the Oseen model using a set of 2×2 matrix operators. His approach should therefore be equivalent to the one given here. However, in the present work, explicit formulae for the rotatory power are given with which earlier work can be compared, and the assumptions and approximations inherent in that work examined.

In this paper a simple exact solution of the Oseen model is presented for light propagating along the z-axis. It can be physically interpreted in terms of a splitting of the dispersion curve (frequency ω vs wave vector K) for light of one circular polarization, similar to the band gap of solid state physics. The expression derived for the optical rotation agrees with that of de Vries well away from λ_0. Near λ_0 it shows anomalous dispersion similar to that derived by CS. Although a self-contained account is given, it is worth mentioning that the method used here was applied earlier[9] to a problem in magnetic neutron diffraction which resembles in many respects the problem of wave propagation in cholesteric liquid crystals.

The second part of the paper deals with the model of CS, with the difference that each birefringent plate, rather than the pitch P, is taken as a unit with associated reflection and transmission coefficients. As in their theory, the amplitudes of the wave satisfy a difference equation. Because we have to consider both polarizations of light simultaneously, these are now matrix difference equations. They are shown to be equivalent to the equations of the Oseen model when we make the plates sufficiently thin and the number per pitch sufficiently large. This calculation unifies the two apparently diverging theoretical approaches and removes the approximations in each.

The methods used in this paper have been applied to the problem of reflection from finite and semi-infinite liquid crystals, and also to a more general model in which the orientation of the molecules varies in a step-like way rather than continuously. The results will be published separately.

2. The Oseen Model

We consider the propagation of light along the z-axis (Fig. 1). The wave equation reads

$$\frac{\partial^2 \mathbf{E}}{\partial z^2} = -\frac{\omega^2}{c^2}\hat{\epsilon}\mathbf{E}. \tag{1}$$

In deriving this, we have used the fact that $\mathbf{E}$ lies in the x-y plane, and have assumed a time dependence $\exp(-i\omega t)$ for it. The tensor $\hat{\epsilon}$ has to be evaluated in the $Oxyz$ system of axes. Since it is given by

$$\begin{bmatrix} \epsilon_a & 0 \\ 0 & \epsilon_b \end{bmatrix}$$

when referred to its own principal axes Oab, which are rotated by qz with respect to Oxy, it is easily calculated to be

$$\hat{\epsilon} = \begin{bmatrix} \epsilon + \beta \cos 2qz & \beta \sin 2qz \\ \beta \sin 2qz & \epsilon - \beta \cos 2qz \end{bmatrix} \tag{2}$$

In (2), $\epsilon = \dfrac{\epsilon_a + \epsilon_b}{2}$

$$\beta = \frac{\epsilon_a - \epsilon_b}{2} = \tfrac{1}{2}(n_a + n_b)(n_a - n_b)$$

$$= \bar{n}\delta n$$

where n_a, n_b are the principal refractive indices in the x-y plane, $\bar{n}$ their average ($n_a{}^2 = \epsilon_a$, $n_b{}^2 = \epsilon_b$) and δn is the birefringence. The tensor in (2) can be regarded as a 2×2 matrix multiplying the 2×1 column matrix $[E_x, E_y]$. We next introduce new variables

$$E_1 = (E_x + iE_y)^{1/2}; \; E_2 = (E_x - iE_y)^{1/2}. \tag{3}$$

The physical significance of the wave described by E_1 is got by putting $E_2 = 0$. We get $E_x = iE_y$, i.e. E_x lags E_y by 90° in phase with our $\exp(-i\omega t)$ convention for the time dependence. This wave is therefore right circular for propagation along $+z$ and we refer to it as the " 1 " wave. The " 2 " wave with $E_1 = 0$ is similarly left circular for propagation along $+z$. This identification is reversed for propagation along $-z$ as the convention for right and left circular polarization makes explicit reference to the direction of propagation, while our convention for " 1 " and " 2 " does not. It is a simple matter to make the substitution (3) into the original wave equation (1) to obtain

$$\begin{bmatrix} \partial^2 E_1/\partial z^2 \\ \partial^2 E_2/\partial z^2 \end{bmatrix} = -\frac{\omega^2}{c^2}\begin{bmatrix} \epsilon & \beta \exp(i2qz) \\ \beta \exp(-i2qz) & \epsilon \end{bmatrix}\begin{bmatrix} E_1 \\ E_2 \end{bmatrix} \tag{4}$$

In crystal optics, it is usual to remove the space dependence by assuming a variation of the form $\exp(ikz)$ for $\mathbf{E}$. It is clear that this fails for a medium in which $\hat{\epsilon}$ depends on z, as in (4). Nevertheless we try a solution of the type $[\exp(ikz), 0]$, i.e., a right circular wave with wave vector k. Substituted into (4), it gives rise only to terms having the same wave vector k on the left side. But the right side equals

$$-\frac{\omega^2}{c^2}\epsilon\begin{bmatrix} \exp(ikz) \\ 0 \end{bmatrix} - \frac{\omega^2}{c^2}\beta\begin{bmatrix} 0 \\ \exp\{i(k-2q)z\} \end{bmatrix}$$

The effect of the dependence of $\hat{}$ on z contained in the β terms of (4), is to convert a wave of the " 1 " polarization into " 2 ", with a shift of wave vector down by $2q$. Similarly using a trial solution of the type $[0, \exp(ikz)]$, we see that it converts a " 2 " wave into a " 1 " wave with an *upward* shift of the wave vector by $2q$. It is clear, then, that a superposition of the type

$$[a \exp\{i(k+q)z\}, \; b \exp\{i(k-q)z\}] \tag{5}$$

is *closed* in the sense that each of the waves appearing in it is converted only into the other, and can therefore satisfy (4) with a proper choice of a and b. Substituting (5) into (4), we get

$$\begin{bmatrix} (k+q)^2 - \dfrac{\epsilon\omega^2}{c^2} & -\dfrac{\beta\omega^2}{c^2} \\ -\dfrac{\beta\omega^2}{c^2} & (k-q)^2 - \dfrac{\epsilon\omega^2}{c^2} \end{bmatrix} \begin{bmatrix} a \\ b \end{bmatrix} = 0. \tag{6}$$

Clearly, the mixing of waves with wave vectors differing by $2q$ is a consequence of Bragg reflection. The only precaution to be observed when using (5) is to remember that the wave vectors are $k+q$ and $k-q$ for the " 1 " and " 2 " waves, but k itself is not the wave vector of any wave.

The condition for (6) to lead to the ratio (a/b) is the vanishing of the determinant.

$$[(k+q)^2 - K_m^2][(k-q)^2 - K_m^2] - \beta^2 K^4 = 0 \tag{7}$$

Here we have introduced $K = \omega/c$, the wave vector in free space, and $K_m = \epsilon^{1/2}\omega/c$ the wave vector corresponding to frequency ω in a (hypothetical) medium of dielectric constant ϵ. (7) is a quadratic in k^2 with the roots

$$k_2, k_1 = [K_m^2 + q^2 \pm (4K_m^2 q^2 + \beta^2 K^4)^{1/2}]^{1/2} \tag{8}$$

The value of a/b corresponding to each of these can be easily got from (6)

$$a/b = \frac{\beta K^2}{(k+q)^2 - K_m^2} = \frac{(k-q)^2 - K_m^2}{\beta K^2} \tag{9}$$

(8) and (9) completely determine the exact solutions of the original equation (1).

We next proceed to study and interpret these solutions. If we put $\beta = 0$ then (8) gives $k_{2,1} = K_m \pm q$. Taking $k_1 = K_m - q$ say, we get from (9) that $a/b = \infty$, i.e., $b = 0$. This is a wave of " 1 " polarization with wave vector $k_1 + q = K_m - q + q = K_m$. Similarly $k_2 = K_m + q$ gives us $a = 0$, a wave of " 2 " polarization with wave vector $K_m + q - q = K_m$. Of course, when $\beta = 0$ the birefringence and with it the inhomogeneity of the medium have disappeared and the normal waves can indeed be chosen as two circular waves with the same wave vector. Considering β to be non-zero has two consequences. From (8), $k_{1,2}$ will differ from $K_m \pm q$ by quantities of order β^2. From (9), it follows for the k_2 solution $a/b \sim \beta^2/\beta \sim \beta$, while for the k_1 solution, $b/a \sim \beta$. We no longer get pure circular waves, but we can continue to refer to the k_1 solution as " 1 " dominant (u_1) and the k_2 solution as " 2 " dominant (u_2). Each normal wave can be described as made up of two oppositely polarized circular waves with wave vectors differing by $2q$, with one of the waves dominating. (5), (8) and (9) are accordingly rewritten

$$\left.\begin{aligned} u_1 &= [\exp(iK_1 z),\ d\exp\{i(K_1 - 2q)z] \\ u_2 &= [f\exp\{i(K_2 + 2q)z\},\ \exp(iK_2 z)] \end{aligned}\right\} \tag{5'}$$

where

$$\left.\begin{aligned} K_1 &= k_1 + q = q + [K_m{}^2 + q^2 - (4K_m{}^2q^2 + \beta^2K^4)^{1/2}]^{1/2} \\ K_2 &= k_2 - q = -q + [K_m{}^2 + q^2 + (4K_m{}^2q^2 + \beta^2K^4)^{1/2}]^{1/2} \end{aligned}\right\} \tag{8'}$$

denote the wave vectors of the dominant components in each case.

$$d = \frac{K_1{}^2 - K_m{}^2}{\beta K^2},\ f = \frac{K_2{}^2 - K_m{}^2}{\beta K^2} \tag{9'}$$

We saw earlier that when β is equal to zero, K_1 and K_2 are both equal to K_m (and, from (9′), $d = f = 0$).

Now K_1 and K_2 are different, the difference being, from (8)

$$\begin{aligned} K_1 - K_2 = {} & [K_m{}^2 + q^2 - (4K_m{}^2q^2 + \beta^2K^4)^{1/2}]^{1/2} \\ & - [K_m{}^2 + q^2 + (4K_m{}^2q^2 + \beta^2K^4)^{1/2}]^{1/2} + 2q \end{aligned} \tag{10}$$

The dominantly right circular and dominantly left circular waves travel with different wave vectors. *This* is the genesis of the optical activity, the optical rotation per unit length being given by $\alpha = (K_1 - K_2)/2$.

Of course the situation is not identical to natural optical activity because the normal waves are not pure circular waves, d and f being non-zero, though small. The effect of the d and f components will cause the polarization of the light, considered, say, on the Poincaré sphere,[10] to deviate from what is predicted on the basis of pure optical rotation. Since these waves have wave vectors differing by $2q$ from those of the dominant components, the resultant motion in Stokes space will have a spatial modulation with this wave vector.

This effect has been considered by Marathay[1] who terms it nutation. We do not consider it further, except to remark that (i) it is implicit even in the rolling cone method used by Mauguin in 1911, where the Stokes vector would execute a wobbling motion; (ii) it is a periodic effect, in contrast to the rotation which is cumulative in z. This accounts for the apparent paradox that we neglect d and f which are of order β, but consider $K_1 - K_2$ which is of order β^2.

The expressions (5′), (8′), (9′) and (10) are exact and describe both total reflection and anomalous rotatory dispersion. For convenience of interpretation and comparison with the de Vries theory, an approximate form of (10) is used. Clearly, from (8′)

$$K_1 - K_2 = k_1 - k_2 + 2q$$

or

$$\begin{aligned}(k_1 - k_2)^2 &= [2q - (K_1 - K_2)]^2 \\ &\simeq 4q^2 - 4q(K_1 - K_2).\end{aligned}$$

Here $K_1 - K_2$ is neglected compared to q, which means that the optical rotation *per pitch* is small compared to 2π.

Therefore

$$-(K_1 - K_2) = \frac{(k_1 - k_2)^2}{4q} - q = \frac{k_1^2 + k_2^2 - 2(k_1^2 k_2^2)^{1/2} - 4q^2}{4q}$$

But the sum $k_1^2 + k_2^2$ and the product of $k_1^2 k_2^2$ are easily got from the quadratic (7) satisfied by k^2.

We get

$$\begin{aligned}-(K_1 - K_2) &= \frac{2(K_m^2 + q^2) - 2[(K_m^2 - q^2)^2 - \beta^2 K^4]^{1/2} - 4q^2}{4q} \\ &= \frac{x - (x^2 - \beta^2 K^4)^{1/2}}{2q}\end{aligned} \quad (11)$$

Here $x = K_m^2 - q^2$.

To deduce the de Vries expression, we must take the further approximation (which is not valid near the Bragg reflection) that $x^2 \gg \beta^2 K^4$.

Then

$$\alpha = \frac{K_1 - K_2}{2} = -\frac{\beta^2 K^4}{8qx} = -\frac{[\bar{n}\delta n]^2 [2\pi/\lambda]^4}{8(2\pi/P)[(2\pi\bar{n}/\lambda)^2 - (2\pi/P)^2]}$$

on substituting for β, K_m, K, x, and q. That is

$$\alpha = \frac{\pi P(\delta n)^2}{4\lambda^2[1-(\lambda^2/\lambda_0^2)]},$$

which is the de Vries expression.

Putting $\lambda \ll \lambda_0$ further gives the expression of Mauguin.(5)

It is clear from (8) or (8′) that if K_m is sufficiently close to q, K_1 can acquire an imaginary part, while K_2 is always real (for positive K_m). Then the value of

$$\alpha = -[x-(x^2-\beta^2K^4)^{1/2}]/4q$$

will also become complex. The real part gives us rotation, and the imaginary part circular dichroism.(10) The width of the reflection band around $K_m = q$ can be estimated with sufficient accuracy by putting $K = K_m/\bar{n} \simeq q/\bar{n}$ in (11). Then α has an imaginary part if $x^2 < q^4\beta^2/\bar{n}^4$, i.e., if $|x| < q^2\beta/\bar{n}^2$.

Since $|x| = |K_m{}^2 - q^2|$, we get total reflection for $K_m = q \pm \Delta K_m$ where $2K_m\Delta K_m \sim 2q\Delta K_m = q^2\beta/\bar{n}^2$ or $\Delta K_m = q\beta/2\bar{n}^2$.

Since

$$\lambda = 2\pi\bar{n}/K_m, \ |\Delta\lambda| = \frac{2\pi\bar{n}}{K_m{}^2}\Delta K_m = \frac{2\pi}{q}\cdot\frac{1}{2}\cdot\frac{\beta}{\bar{n}} = \frac{P\delta n}{2}.$$

The width of the region of reflection as referred to free-space wavelength is $2|\Delta\lambda| = P\delta n$ subject to the approximations made. A more accurate expression can easily be got from (8).

We have assumed that the attenuation of the " 1 " wave shown by the imaginary part of K_1 is associated with total reflection of that wave. This is clear from the fact that there is no dissipative mechanism built into the model. We can also study the reflection by considering the coefficient d, whose value is given by (9′). Putting $K_m = q$ (at the centre of the reflection band)

$$K_1 = q + [2q^2 - (4q^4 + \beta^2q^4/\bar{n}^4)^{1/2}]^{1/2}$$
$$\simeq q + iq^2\beta/2\bar{n}^2$$
$$d = \frac{(K_1{}^2 - q^2)\bar{n}^2}{\beta q^2} \simeq i.$$

Clearly, as λ approaches λ_0, $|d|$ becomes of order 1. This qualifies our earlier statement that d and f are small quantities. An exact treatment of reflection from a finite slab would be to match an incoming and reflected wave on one side of the slab to four waves within the slab (two in the forward and two in the backward direction) and a transmitted wave on the other side.

3. Equivalence of the Continuum and Dynamical Approaches

We now consider the same model as CS. The liquid crystal is represented by an array of birefringent plates with mean refractive index $(n_a+n_b)/2 = \bar{n}$ and birefringence $(n_a-n_b) = \delta n$ where n_a and n_b are the two principal refractive indices. Let τ be the thickness of the plate. The optical properties of each plate are fully specified by r_a, t_a, r_b and t_b, the reflection and transmission coefficients for light linearly polarized along the two principal directions at normal incidence. In fact, these can be obtained by substituting n_a and n_b into the well-known expressions for r and t in terms of n, the refractive index of an isotropic plate.

$$r = \frac{(n^2-1)(1-p^2)}{(n+1)^2-(n-1)^2p^2}$$

$$t = \frac{4np}{(n+1)^2-(n-1)^2p^2}\ ; \ p \equiv \exp(inK\tau) \quad (12)$$

K is the wave vector in vacuum.

We will need an identity satisfied by r and t. If $t = |t|\exp(i\gamma)$ then it can be shown that

$$t^2 - r^2 = \exp(2i\gamma) \quad (13)$$

Further, only the limiting case of small τ is of interest. For this,

$$|t| = 1 - \frac{(n^2-1)^2}{8}K^2\tau^2$$

$$\gamma = \left(\frac{n^2+1}{2}\right)K\tau \quad (14)$$

In order to set up the difference equations for this model, we first consider a single plate and define E_n and e_n, the amplitudes emerging from the n^{th} plate in the positive and negative z directions respectively. I_n and i_n are, again, the amplitudes incident on the n^{th} plate in the positive and negative z directions respectively (Fig. 2).

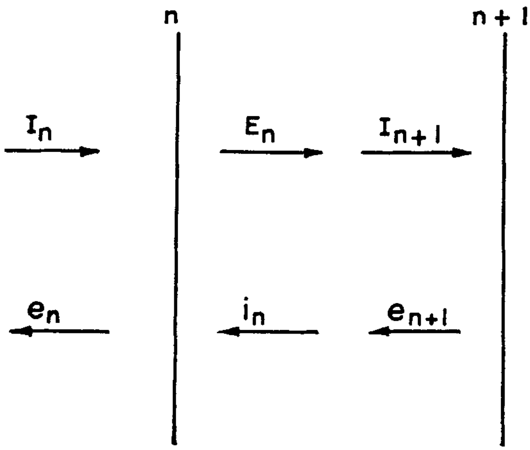

Figure 2. The symbols used in setting up the difference equations of the dynamical theory. The arrow associated with each shows the direction of the corresponding wave.

E_n is made up of the transmitted part of I_n and the reflected part of i_n:

$$\left.\begin{aligned} E_n &= tI_n + ri_n \\ e_n &= rI_n + ti_n \end{aligned}\right\} \tag{15}$$

The second equation in (15) is derived in the same way as the first. Inverting (15)

$$\left.\begin{aligned} (t^2 - r^2)I_n &= tE_n - re_n \\ (t^2 - r^2)i_n &= -rE_n + te_n \end{aligned}\right\} \tag{16}$$

In Eqs. (15) and (16), we can regard all the quantities E, e, I and i as referring to a wave polarized along one of the principal axes (say a) of the plate, in which r and t must take the values (r_a and t_a) referring to that axis. We can combine the equations referring to the a and b axes in the following way

$$\begin{bmatrix} E_n^a \\ E_n^b \end{bmatrix} = \begin{bmatrix} t_a & 0 \\ 0 & t_b \end{bmatrix} \begin{bmatrix} I_n^a \\ I_n^b \end{bmatrix} + \begin{bmatrix} r_a & 0 \\ 0 & r_b \end{bmatrix} \begin{bmatrix} i_n^a \\ i_n^b \end{bmatrix} \tag{17}$$

for the first line of (15).

We will continue to write this equation in the form $E_n = tI_n + ri_n$, with the understanding that from now onwards E, I, e, i are column vectors (2×1 matrices) and r and t are 2×2 diagonal matrices.

The convenient form of r and t arises from the choice of E_n^a and E_n^b along the principal axes of the n^{th} plate. The emergent wave E_n from the n^{th} plate is physically the same as I_{n+1}, viz., that incident on the $(n+1)^{\text{th}}$ plate. However (15) and (16) will only apply to the plate if I_{n+1} is referred to *its* principal axes, which are rotated by an angle θ from the axes used to express E_n. We write, therefore, $I_{n+1} = SE_n$ and similarly $i_n = S^{-1}e_{n+1}$, where

$$S = \begin{bmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{bmatrix} \tag{18}$$

The basic difference equations are now easily derived from Eqs. (15) to (18):

$$(t^2 - r^2)I_{n+1} = tE_{n+1} - re_{n+1} \qquad \text{[from (16)]}$$

$$= tE_{n+1} - rSi_n \qquad \text{[from (18)]}$$

$$= tE_{n+1} - rSr^{-1}(E_n - tSE_{n-1}) \qquad \text{[using (15) and (18)].}$$

Premultiplying by S^{-1}

$$S^{-1}(t^2 - r^2)SE_n = S^{-1}tE_{n+1} - S^{-1}rSr^{-1}E_n + S^{-1}rSr^{-1}tSE_{n-1}.$$

This equation can be considerably simplified in the limiting case of interest $\tau \to 0$, $\theta \to 0$, $\theta/\tau = q$. That is, the angle of twist between the plates is made smaller and the plates thinner, maintaining the same pitch. Only in this case can one look for equivalence with the continuum model. (The effect of a discrete structure is to be considered in another paper.) We therefore retain only the lowest power of τ which appears. A study of the previous equation shows that it is τ^2, so that we are justified in neglecting a term like $\theta\tau^2$ (which is of order τ^3). In a product like $S^{-1}rSr^{-1}$ we can write the inner rS as $Sr + (rS - Sr)$ and neglect the term $(rS - Sr)$ which when evaluated is seen to be of a higher order. Similarly we can write $S^{-1}t$ as tS^{-1}. Making these changes, the difference equation takes the form $(t^2 - r^2)E_n + E_n = t(S^{-1}E_{n+1} + SE_{n-1})$.

Using (13),

$$[\exp(2i\gamma) + 1]E_n = t\exp(i\gamma)(S^{-1}E_{n+1} + SE_{n-1}),$$
$$2\cos\gamma\, E_n = t(S^{-1}E_{n+1} + SE_{n-1}) \tag{19}$$

We may recall at this point that $2 \cos \gamma$ is really a matrix

$$\begin{bmatrix} 2\cos\gamma_a & 0 \\ 0 & 2\cos\gamma_b \end{bmatrix},$$

and so is $|t|$.

As before, we introduce new variables

$$E'_{1,2} = (E^a \pm iE^b)/2^{1/2}.$$

The matrices occurring above have to be transformed using the matrix

$$A \equiv 2^{-1/2}\begin{bmatrix} 1 & i \\ 1 & -i \end{bmatrix}.$$

For example

$$S' = ASA^{-1} = \begin{bmatrix} \exp(-i\theta) & 0 \\ 0 & \exp(i\theta) \end{bmatrix}$$

and

$$r' = \begin{bmatrix} \dfrac{r_a + r_b}{2} & 0 \\ 0 & \dfrac{r_a + r_b}{2} \end{bmatrix} + \begin{bmatrix} 0 & \dfrac{r_a - r_b}{2} \\ \dfrac{r_a - r_b}{2} & 0 \end{bmatrix}.$$

In what follows we write

$$\frac{r_a + r_b}{2} = \bar{r}, \qquad \frac{r_a - r_b}{2} = \frac{\delta r}{2}.$$

Further, because of the periodicity with respect to n, the difference equation can be solved in the form

$$E_n' = E' \exp(in\phi) \tag{20}$$

A real ϕ implies an unattenuated solution and a complex ϕ an attenuated solution. Making the change of variables and using (20), we get

$$\begin{aligned} &\left\{2\cos\bar{\gamma} + \begin{bmatrix} 0 & \sin\gamma\delta\gamma \\ \sin\bar{\gamma}\delta\gamma & 0 \end{bmatrix}\right\} E' \exp(in\phi) \\ &= \left\{|\bar{t}| + \tfrac{1}{2}\begin{bmatrix} 0 & \delta|t| \\ \delta|t| & 0 \end{bmatrix}\right\} \\ &\quad \times \left\{\begin{bmatrix} \exp(i\theta) & 0 \\ 0 & \exp(-i\theta) \end{bmatrix} E' \exp[i(n+1)\phi] \right. \\ &\quad \left. + \begin{bmatrix} \exp(-i\theta) & 0 \\ 0 & \exp(i\theta) \end{bmatrix} E' \exp[i(n-1)\phi]\right\} \end{aligned} \tag{21}$$

As mentioned earlier, we are working to second order in τ. So we approximate $\cos\gamma$ by $1-\frac{1}{2}\gamma^2$ [since γ is of the first order in τ from (14)] and make similar approximations for $\cos(\phi\pm\theta)$. We then get, from (21)

$$\begin{bmatrix} -\bar{\gamma}^2+|\bar{t}|\cos(\phi+\theta) & \bar{\gamma}\delta\gamma-\delta|t| \\ \bar{\gamma}\delta\gamma-\delta|t| & -\bar{\gamma}^2+|\bar{t}|(\phi-\theta)^2 \end{bmatrix}\begin{bmatrix} E_1' \\ E_2' \end{bmatrix}=0.$$

From (14) $\delta\gamma=\bar{n}\,\delta nK\tau$,

$$\delta|t|=\frac{\bar{n}(\bar{n}^2-1)}{2}\,\delta nK^2\tau^2$$

Using these results and the values of $\bar{\gamma}$ and $\bar{t}$ from (14) we get

$$\begin{bmatrix} \left(\dfrac{\phi+\theta}{\tau}\right)^2-\bar{n}^2K^2 & \bar{n}\delta nK^2 \\ \bar{n}\delta nK^2 & \left(\dfrac{\phi-\theta}{\tau}\right)-\bar{n}^2K^2 \end{bmatrix}\begin{bmatrix} E_1' \\ E_2' \end{bmatrix}=0. \qquad (22)$$

Equation (22) is equivalent to Eq. (6) when we identify ϕ/τ with k, θ/τ with q, and $\epsilon\omega^2/c^2$ with $\bar{n}^2K^2$, i.e., $K_m{}^2$.

All the deductions from (6) are also valid for this model. In this way, we have shown the dynamical theory as applied to the pile of birefringent plates to be equivalent to the Oseen model. Taking the basic structural unit in the difference equation to be of thickness P and the properties of this unit to be described at all wavelengths by the Mauguin formula seems to be the main approximation in the CS treatment which is responsible for the slight discrepancy in their theory at very long wavelengths. We may conclude that if a second change in the sign of rotation is observed at $\lambda \gg \lambda_0$, it must be attributed to a different structure or to another contribution to the rotation superposed on the one we have considered.

4. An Illustrative Example

The results of calculations based on (5′), (8′), (9′) and (10) are shown in Fig. 3. In Fig. 3(a), the value of K_2 and the real and imaginary parts of K_1 are plotted as functions of the wavelength in free space λ. The parameters chosen are $n_a = 1.4$, $n_b = 1.47$, $P = 3571$ Å, which gives rise to a reflection band near 5000 Å, of width 250 Å. We

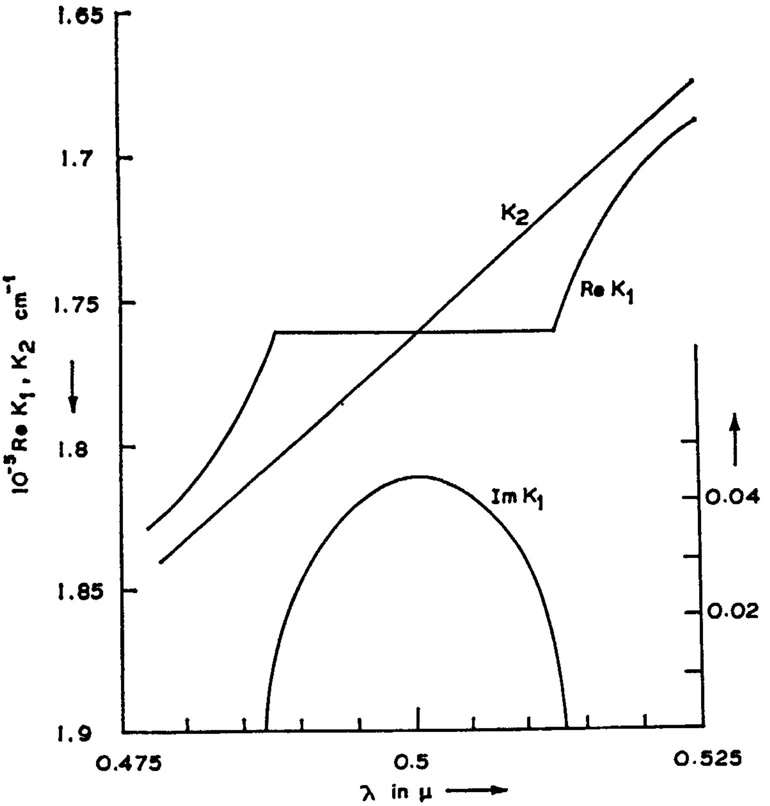

Figure 3(a). The variation of K_1 and K_2, the dominant wave vectors of the two normal waves, plotted as functions of the free space wavelength λ.

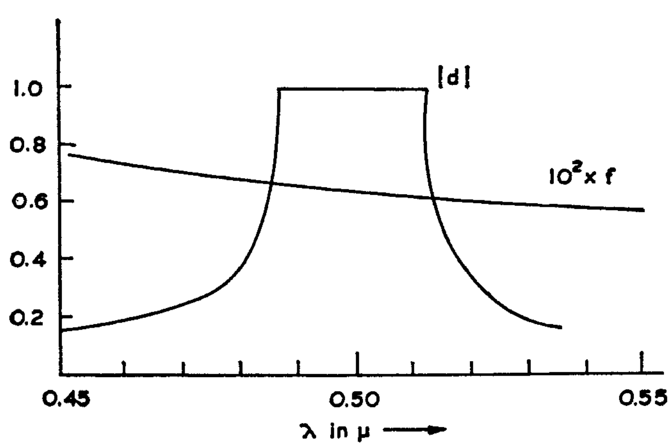

Figure 3(b). The coefficients d and f, which describe the mixed character of the normal waves, as functions of λ.

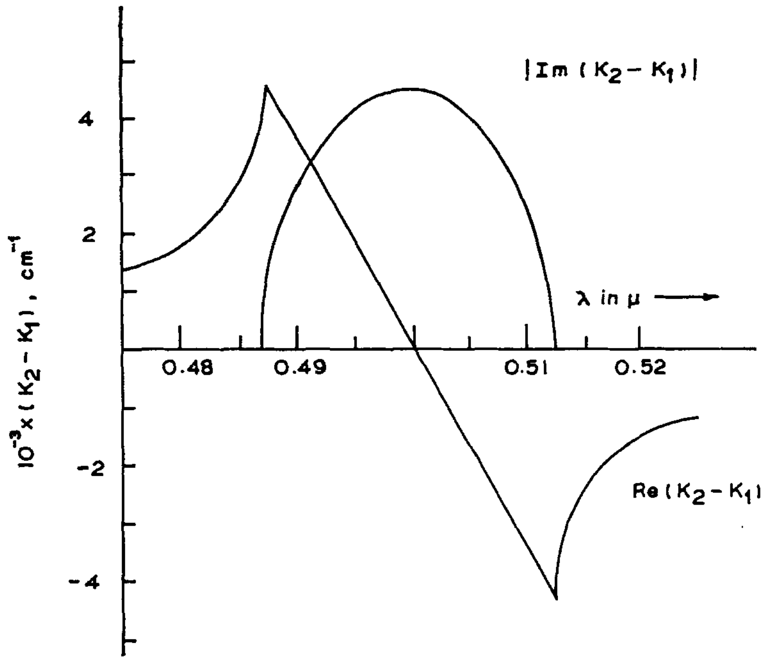

Figure 3c. The real and imaginary parts of $(K_2 - K_1)$, i.e., optical rotation and circular dichroism respectively, as functions of λ.

neglect the variation of n_a and n_b with wavelength. The gap in the curve for the real part of K_1 is seen. Figure 3(b) plots $|d|$ in the same region of wavelengths, and shows the growth of $|d|$ as we approach λ_0. In the same region f is small, showing that the " 2 " wave does not undergo Bragg reflection. In the band, $|d| = 1$ so that we have a standing wave, which is of course attenuated by the imaginary part of K_1. Figure 3(c) shows the real and imaginary parts of $K_2 - K_1$ which represent the rotation and circular dichroism.

5. Conclusions

The Oseen model is seen to admit of an exact solution. The two vital steps in the derivation are (i) the introduction of circularly polarized waves as basis—which is natural considering the symmetry of the structure, (ii) assuming a superposition of waves of *opposite* circular polarizations and *different* wave vectors related by the Bragg condition as a trial solution. The circumstance which makes an exact solution possible is that each component of the solution is only scattered into the other, and together they form a closed set from which a true normal wave can be built up. (It is the absence of such a closed property which makes an analytical solution difficult for propagation off the axis.) The normal waves are not circular waves.

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REFERENCES

1. Oseen, C. W., *Trans. Faraday Soc.* **29**, 833 (1933).
2. Vries, H. de, *Acta Cryst.* **4**, 219 (1951).
3. Darwin, C. G., *Phil. Mag.* **27**, 315, 675 (1914).
4. Chandrasekhar, S. and Srinivasa Rao, K. N., *Acta Cryst.* **A24**, 445 (1968).
5. Mauguin, M. C., *Bull. Soc. Franc. Miner. Crist.* **34**, 71 (1911).
6. Chandrasekhar, S. and Shashidhara Prasad, J., *Mol. Cryst. and Liq. Cryst.* **14**, 115 (1971).
7. Aihara, M. and Inaba, H., *Optics Comm.* **3**, 77 (1971).
8. Marathay, A. S., *Optics Comm.* **3**, 369 (1971); *J. Opt. Soc. Amer.* **61**, 1363 (1971).
9. Rajaram Nityananda and Ramaseshan, S., *Sol. State Comm.* **9**, 1003 (1971).
10. Ramachandran, G. N. and Ramaseshan, S., *Handbuch der Physik*, Vol. XXV/1, Springer Verlag, Berlin (1961).

Note Added in Proof

After submitting this paper, I came across the work of E. I. Kats (*Zh. Eksperim. i. Teor, Fiz.* **59**, 1854 (1970), *Sov. Phys. JETP* **32**, 1004 (1971) in which the results of the first part of this paper, pertaining to the Oseen model, have been anticipated. The method used is identical to that given here and in Ref. 9. In addition, his paper gives an approximate solution of the oblique incidence problem. However, it is stated there that the pile of plates model leads to incorrect results. The second part of this paper shows that this is not the case.