CHAPTER III

COMPARISON OF THE DYNAMICAL THEORY AND THE REGOROUS ELECTROMAGNETIC THEORY OF LIGHT PROPAGATION ALONG THE HELICAL AXIS OF A CHOLESTERIC LIQUID CRYSTAL

1. Introduction

We have so far considered the optical properties of a cholesteric liquid crystal of pitch much larger than the wavelength of light. In this Chapter and in the following one we shall be conaerned with the situation in which the pitch is comparable to the wavelength of light. It is in this region that a cholesteric exhibits some of its distinctive properties. In a plane textured cholesteric, for normal incidence, circularly polarised light having handedness same as that of the helix undergoes selective reflection over a small region of the spectrum. The sense of the reflected light remains the same as that of the incident light. Along the helix the medium has an optical. rotation of the order of thousands of degree per mm. and the rotation shows an anomaly in the neighbourhood

of the reflection band. Also, at the reflection band the medium exhibits circular dichroism.

The general eleotromagnetic theory of light propagation along the optic axis of a cholesteric was formulated by Mauguin (1911), Oseen (1933) and de Vries (1951). Exact solutions based on the spiralling dielectric ellipsoid model have been worked out. This theory haa since been presented in various forms by other authors (Joly 1972, Aihara and Inaba 1971, Marathay 1971), the most recent treatment being that derived by Kats (1971) and independently by Nityananda (1973). Though these rigorous treatments are valid for any arbitrary thickness of the sample and for the entire range of wavelengths, calculations for any practical situation are rather tedious and require the **use** of a computer. An extremely elegant approach to this problem which leads to simple analytical expressions for the reflection coefficient, rotatory power and circular dichroism was proposed by Chandraeekhar and Srinivasa Bao (1968), based on an analogy with Darwin's dynamical theory of X-ray diffraction (1914, 1922). It is the aim of this Chapter to present this theory in complete detail (avoiding certain inconsistencies that were present in the original formulation) and to compare its predictions with those of the exact electromagnetic treatment. Theoretical curves will be presented for the reflection coefficient, anomalous rotatory power, etc. for thick and thin samples and it will be shown that the simple dynamical approach is sufficient for most practical problems.

2. Dynamical theory of cholesteric liquid crystals

It has been shown in Chapter I that when $\beta \gg \gamma$ (i.e., $\frac{P \Delta \mu}{2} \ll 1$), the cholesteric liquid crystal can be considered as a pure rotator. In such a case, right and left circular vibrations travel without change of form but at slightly different velocities. Now, in a pure rotator which rotates linearly polarised light through an angle a per unit thickness, the right- and left-circularly polarised waves undergo phase changes of φ_0 + a and φ_0 - a respectively, where $\mu_{\mathbf{R}}$ and $\mu_{\mathbf{L}}$ are the refractive indices for right and left; circularly polarised wave in **the** medium.

(a) Kinematical theory of reflection

In this approximation one ignores multiple reflections from the layers. Let the principal axes of the first layer be along OX, OX of a Cartesian ooordinate system and let; the structure be right handed, i.e., β is positive. Right circularly polarised light given by $D_0 = \begin{bmatrix} 1\\ 1\\ 1 \end{bmatrix}$ referred to OX, OX is incident; along OZ. The incident light vector is resolved along the principal axes of the $(\gamma + 1)^{\text{th}}$ layer which are inclined at an angle $(\gamma' + 1)\beta$ with respect to OX, OY. The resolved oomponents are

$$\begin{bmatrix} \zeta \\ \eta \end{bmatrix} = \begin{bmatrix} 1 \\ 1 \\ i \end{bmatrix} \exp[i\{(\nu + 1)\beta - \varphi_{\nu+1}\}]$$

where
$$\varphi_{\gamma+1} = \frac{2\pi \mu_R(\gamma+1)b}{\lambda}$$

At the boundary between $(\mathcal{V} + 1)^{\text{th}}$ and $(\mathcal{V} + 2)^{\text{th}}$ layers, the ζ vibration emerges from a medium of refractive index μ_1 and η vibration from a medium of refractive index μ_2 . If ζ' and η' refer to the principal axes of the $(\mathcal{V} + 2)^{\text{th}}$ layer, then the reflected components are

$$\begin{bmatrix} \zeta' \\ \eta' \end{bmatrix} = -\frac{\beta \Delta \mu}{2\mu} \begin{bmatrix} 1 \\ 1 \end{bmatrix} \exp[1\{(\gamma + 1)\beta - \varphi_{\gamma+1}\}]$$

=
$$-iq \begin{bmatrix} 1 \\ -i \end{bmatrix} exp[i\{(\gamma+1)\beta - \varphi_{\gamma+1}\}]$$

where $|\mathbf{q}| = \frac{\beta \Delta \mu}{2\mu}$ is the reflection coefficient of one layer. Since $\beta = \frac{2\pi b}{P}$ ($\mathbf{p} \sim 10$ Å, $P \sim 5000$ Å) is small, $\sin \beta \approx \beta$. On reflection there occurs a slight ellipticity in the beam which is neglected.

Transforming back to OX, OX the reflected wave on **reaching** the surface of the liquid crystal will be

$$\begin{bmatrix} \mathbf{X} \\ \mathbf{Y} \end{bmatrix} = -\mathbf{i}\mathbf{q} \begin{bmatrix} \mathbf{1} \\ -\mathbf{i} \end{bmatrix} \exp[\mathbf{i}\{(2\mathbf{y} + 3)\mathbf{\beta} - 2\mathbf{\varphi}_{\mathbf{y}+1}\}]$$

which represents a right circular vibration travelling in the negative direction of OZ. The phase difference between this wave and that reflected at the boundary between first an8 second layer is $[2(\gamma\beta - \varphi_{\gamma})]$.

When $\lambda =$, the phase

$$\varphi_1 = \frac{2\pi\mu_R p}{\lambda} = \frac{2\pi}{n} = \beta$$

and

Hence the phase factor $\exp[2i(\vartheta\beta - \varphi_{\vartheta})]$ becomes unity irrespective of the value of \mathcal{V} , and there results a strong interference maximum. On the other hand, for a left-handed structure, β is negative and $(\vartheta\beta - \varphi_{\vartheta}) \neq 0$ when $\lambda = \mu_R P$. Therefore the waves from the different layers will not be in phase and recults in total transmission. Thus when the wavelength of light in the medium is equal to the pitch of the sample, reflection of one of the circular component which has the same sense as that of the helix takes place and contrary to reflection from a dielectric medium, the reflected wave has the same sense of circular polarisation as that of the incident wave.

In the kinematical approximation the reflection ooefficient per turn of the helix ie

$$-iQ = -inq = -i\pi \frac{\Delta \mu}{\mu} \qquad (2)$$

b) Dynamical theory of reflection

So far only one reflection from each layer has been considered and secondary reflections have been neglected. In the kinematical approximation considered above, multiple reflections from the repetitive units constituting the structure hare been ignored, but the complete solution of the problem has to take this into account. This is the essence of the dynamical theory.

Bor thie purpose, let us regard the liquid orystal as **consisting** of a set of parallel planes **spaced** P apart. Each plane **therefore replaces** the 'n' **birefringent layers** per turn of the helix of **pitoh** P. Let the reflection coefficient per plane be -iQ for right circular light at normal **incidence**. Assuming the kinematical approximation for the **n layers**, Q is given by (2).

Let $\mathbf{T_r}$ and $\mathbf{S_r}$ be the **complex** amplitudes of the primary and reflected waves st a point **just** above the $\mathbf{r^{th}}$ plane, the **topmost** plane being **designated** by the serial number **zero** (**Figure** 1). Neglecting absorption, the difference equation may be written as



<u>Picture 1</u>: Reflections from a set of parallel planes spaced at a distance P apart. T_r and S_r represent the complex amplitudes of the primary and reflected waves respectively at a point just above the rth plane.

$$S_{r} = -iQT_{r} + exp(-i\varphi)S_{r+1}$$
(3)

$$\mathbf{T}_{r+1} = \exp(-i\varphi)\mathbf{T}_r - iQ \exp(-2i\varphi)\mathbf{S}_{r+1}$$
(4)

where $\varphi = \frac{2\pi \mu_R P}{\lambda}$. The reflection coefficient here is taken to be the same on both sides of the plane. Replacing r by (r = 1) in (3) and (4), substituting and simplifying, it leads to

$$\mathbf{T}_{r+1} + \mathbf{T}_{r-1} = \mathbf{Y} \mathbf{T}_r \tag{5}$$

$$S_{r+1} + S_{r-1} = Y S_r$$
 (6)

where

$$X = \exp(i\varphi) + \exp(-i\varphi) + Q^2 \exp(-i\varphi)$$
 (7)

Supposing the film to consist of m planes and if $S_m = 0$, then from (6)

$$S_{m-2} = Y S_{m-1}$$
,
 $S_{m-3} = Y S_{m-2} - S_{m-1} = (Y^2 - 1)S_{m-1}$,

non 44

$$S_{m-4} = (Y^3 - 2Y)S_{m-1}$$
, etc.,

and

$$S_{o} = [Y^{m-1} - (\underline{m-2}) Y^{m-3} + (\underline{m-4})(\underline{m-3}) Y^{m-5} - ...]S_{m-1}$$
$$= f_{m}(y)S_{m-3} (say) (8)$$

Similarly from (4), (5) and (7)

$$T_{m-1} = \exp(i\varphi)T_m$$

$$T_{m-2} = [y \exp(i\varphi) - 1]T_m$$

$$T_{m-3} = [(y^2 - 1)\exp(i\varphi) - y]T_m, \text{ etc.}$$

and

$$\mathbf{T}_{o} = [\mathbf{f}_{m}(\mathbf{y})\exp(\mathbf{i}\boldsymbol{\varphi}) - \mathbf{f}_{m-1}(\mathbf{y})]\mathbf{T}_{m}$$
(9)

Since from (3),

$$S_{m-1} = -iQT_{m-1} = -iQexp(i\phi)T_m$$
,

the ratio of the reflected to incident amplitude is

$$\frac{S_{o}}{T_{o}} = \frac{-iQ f_{m}(y) \exp(i\varphi)}{f_{m}(y)\exp(i\varphi) - f_{m-1}(y)}$$
(10)

A relation in the form $T_{r+1} = xT_r$ is assumed so that x satisfies

$$\mathbf{x} + \frac{1}{\mathbf{x}} = \mathbf{y} = \exp(\mathbf{i}\varphi) + \exp(-\mathbf{i}\varphi) + Q^2 \exp(-\mathbf{i}\varphi) \quad (10')$$

The reflection condition is $\mu_R P = \lambda_0$ or $\varphi_0 = 2\pi$. Accordingly one may writa,

$$\varphi = \frac{2\pi\lambda_0}{\lambda} = \varphi_0 + \varepsilon,$$

where

$$\varepsilon = -\frac{2\pi(\lambda - \lambda_o)}{\lambda}$$

which is a small quantity in the neighbourhood of the reflection band. Therefore,

$$x + \frac{1}{x} = \exp(i\varepsilon) + \exp(-i\varepsilon) + Q^2 \exp(-i\varepsilon)$$
(11)

,

This suggests that in the neighbourhood of the

reflection band one may put

$$\mathbf{x} = \exp(-\xi) \exp(-i\varphi_0) = \exp(-\xi) \qquad (12)$$

where ξ is small and may be complex. From (11) and (12)

$$\xi = \pm \left(\varrho^2 - \varepsilon^2 \right)^{\frac{1}{2}} -$$

When

$$y = e \times \xi$$
) + $exp(-\xi) = 2cosh \xi$,

the series in (8) is given by

$$f_{m}(y) = \frac{\sinh m \xi}{\sinh \xi}$$
(13)

Substituting in (10) and simplifying

$$\frac{S_{o}}{T_{o}} \approx - \frac{iQ}{i\varepsilon + \xi \coth m \xi}$$
(14)

Or

$$R = \left| \frac{s_o}{T_o} \right|^2 = \frac{q^2}{\epsilon^2 + \xi^2 \coth^2 m \xi}$$
(15)

From (9) and (13)

$$\frac{\mathbf{T}_{\mathrm{m}}}{\mathbf{T}_{\mathrm{o}}} = \left[\exp(\mathrm{i}\varepsilon) \frac{\sinh \mathrm{m}\xi}{\sinh \xi} - \frac{\sinh (\mathrm{m}-1)\xi}{\sinh \xi} \right]^{-1}$$

$$\approx \frac{\xi \text{ ooeeuh } \mathfrak{m} \xi}{\mathfrak{i}\varepsilon + \xi \operatorname{coth } \mathfrak{m} \xi}$$
(16)

Thue

$$\left|\frac{\mathbf{T}_{m}}{\mathbf{T}_{o}}\right|^{2} + \left|\frac{\mathbf{S}_{o}}{\mathbf{T}_{o}}\right|^{2} = 1$$

For a thick specimen, $\mathbf{m} = \mathbf{\infty}$

$$\frac{S_o}{T_o} = -\frac{Q}{\varepsilon + i\xi}$$
(17)

When $-Q \langle \varepsilon \langle Q, is real$ $R = \left| \frac{S_0}{T_0} \right|^2 = 1$

The reflection is total within this range. The epectral width of the total reflection $\Delta \lambda = \frac{Q\lambda}{\pi}$ $\approx \frac{Q\lambda_0}{\pi}$. Using (2) one gets $\Delta \lambda = P \Delta \mu$, in agreement with the de Vries theory (1951).

48

c) The anomalous rotatory dispersion

If multiple reflections are neglected, the optical rotation per pitch \mathbb{P} of the liquid crystal is $\frac{1}{2}(\varphi_R - \varphi_L)$ and the rotatory power is given by (1). Near the region of reflection, the right circular component suffers anomalous phase retardation and under certain circumstances attenuation as it travels through the medium. Left circular light on the other hand exhibits normal behaviour throughout and as a consequence the rotatory dispersion is anomalous around the reflection region.

Thick specimen:

From (10') and (12)

$$\mathbf{T}_{\mathbf{r}+1} = \mathbf{x} \mathbf{T}_{\mathbf{r}}$$

where

$$x = \exp(-\xi) \exp(-i\varphi_0)$$

$$\xi = \pm (Q^2 - \varepsilon^2)^{\frac{1}{2}}$$

$$\varphi_0 = \varphi_R - \varepsilon = 2\pi$$

Inside the totally reflecting range, ξ is real and therefore the medium becomes highly circularly dichroic. If very thin films are employed, the emergent light is elliptically polarised. It is readily seen that the ellipticity ' χ ' produced per thickness P is given by

$$\tan \chi = \frac{1 - \exp(-\xi)}{1 + \exp(-\xi)} = \tanh \frac{\xi}{2}$$

OT

λ

The azimuth of major **axis** of the ellipse after **passing** through a **thickness P** is

$$\alpha = \frac{1}{2}(\varphi_{R} - \varphi_{I}) = \frac{\pi P}{\lambda}(\mu_{R} - \mu_{I}) + \frac{\pi(\lambda - \lambda_{0})}{\lambda}$$
$$= -\frac{\pi \gamma^{2}}{2\beta} + \frac{\pi(\lambda - \lambda_{0})}{\lambda}$$
Here $\gamma = \frac{\pi P(\Delta \mu)}{\lambda}$. Therefore the rotatory power

$$S = -\frac{\pi(\Delta \mu^2)P}{4\lambda^2} + \frac{\pi(\lambda - \lambda_0)}{P\lambda}$$
(18)

50

-

which is valid within the range

$$(\lambda_{o} - Q/2\pi) \langle \lambda \langle (\lambda_{o} + Q/2\pi) \rangle$$
.

Outside the totally reflecting range $\xi = i(\epsilon^2 - q^2)^{\frac{1}{2}}$ and may be positive or negative depending on whether ϵ is **positive** or negative. Therefore.

$$\alpha = \frac{1}{2} [(\epsilon^2 - Q^2)^{\frac{1}{2}} + \varphi_0 - \varphi_L]$$
$$= -\frac{n\gamma^2}{2\beta} - \frac{\epsilon}{2} [1 - (1 - \frac{Q^2}{\epsilon^2})^{\frac{1}{2}}]$$

Hence the rotatory power

$$S = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2} + \frac{\pi(\lambda - \lambda_0)}{P\lambda} \left[1 - (1 - \frac{Q^2}{\epsilon^2})^{\frac{1}{2}}\right] \quad (19)$$

[Here we follow the **sign** convention that a **clock**wise rotation as seen by an observer looking in the direction of propagation of light to be positive. This is opposite to the **sign** convention followed usually in experiments where a clockwise rotation as seen by an observer looking at the source is taken to be **positive**.]

Thin film

For a thin film the phase of the right circular wave after passing through 'm' planes can be evaluated from (16)

$$\frac{T_{m}}{T_{o}} = A \exp[-im(\varphi_{o} + \gamma)]$$
(20)

where

$$\tan \mathbf{m} \gamma = \frac{\varepsilon}{\xi \coth \mathfrak{m} \xi}$$
(21)

The optical rotation for thickness P 18

$$\frac{1}{2}(\varphi_{0} + \gamma - \varphi_{L}) = \frac{1}{2}[(\varphi_{R} - \varphi_{L}) + (\gamma - \varepsilon)]$$

and the rotatory power

$$S = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2} + \frac{(\gamma - \varepsilon)}{2P} \qquad (22)$$

One can see that the **dynamical** theory explains all the optical properties (within and in the neighbourhood of λ_0) of a plane texture cholesteric liquid crystal. This theory is elegant and leads to simple formulae for optical rotatory power, intensity of the reflected wave, intensity of the transmitted wave in the case of thin as well as semi-infinite cholesteric samples. A brief review of the exact electromagnetic theory as treated by Nityananda (1973) is presented below and then the dynamical theory is compared with it.

3. Exact electromagnetic theory

This is based on Oseen's (1911) model wherein the liquid crystal is treated a8 continuously twisted anisotropic dielectric. Locally this can be described by a dielectric tensor with principal axes ca and ob and principal values ε_a and ε_b . As one moves along the Z-axis (say), oa and ob rotate in the x-y plans through an angle QZ. The pitch P is the distance along the Z-axis corresponding to a rotation of 2π , so that: $P = 2\pi/q_b$.

The wave equation for light propagation along the Z-axis can be derived from Maxwell's equations and it is of the form

$$\frac{\partial^2 \tilde{E}}{\partial z^2} = -\frac{w^2}{c^2} \tilde{\epsilon} \tilde{E} \qquad \left[w = \frac{2\pi c}{\lambda}\right] \qquad (23)$$

[Here a parameter with a bar represents a veotor and that with a tilde represents a tensor.]

where **E** lies in the X-X plane and it is assumed to have a time dependence exp(-iwt).

is a tensor and is given by



Transforming it to oab axes, which is rotated through an angle $q_0 Z$ with respect to OXY,

where $\begin{bmatrix} \varepsilon + B \cos 2q_0 Z & B \sin 2q_0 Z \\ B \sin 2q_0 Z & \varepsilon = B \cos 2q_0 Z \end{bmatrix}$ (24)

$$E = \frac{e_{a} - e_{b}}{2} = \frac{1}{2}(\mu_{a} + \mu_{b})(\mu_{a} - \mu_{b})$$
$$= \mu \delta \mu .$$

Here μ_a , μ_b ore the principal refractive indices in the X-Y plane ($\mu^2 = \epsilon_a$, $\mu^2_b = \epsilon_b$) = refractive index μ is the average and $\delta\mu$ is the birefringence.

Let us introduce two new variables E_1 and E_2 defined by

$$E_{1} = \frac{(E_{x} + iE_{y})}{(2)^{\frac{1}{2}}},$$

$$E_{2} = \frac{(E_{x} - iE_{y})}{(2)^{\frac{1}{2}}}.$$
(25)

One can see the physical significance of E_1 by putting $E_2 = 0$. Then $E_x = iE_y$, i.e., E_x lags E_y by 90° in phase with the exp^{-iwt} convention for time dependence. Therefore E_1 , with $E_2 = 0$, represents right circular wave for propagation along +2. This wave is referred re '1' wave. Similarly with $E_1 = 0$, E_2 represents left circular wave for propagation along -2. This is referred re '2' wave.

Substituting equation (25) in (23) gives

$$\begin{bmatrix} \partial^2 \mathbf{E}_1 / \partial \mathbf{Z}^2 \\ \partial^2 \mathbf{E}_2 / \partial \mathbf{Z}^2 \end{bmatrix} = -\frac{\mathbf{w}^2}{\mathbf{o}^2} \begin{bmatrix} \mathbf{\varepsilon} & \operatorname{Bexp}(\mathbf{i} 2\mathbf{q}_0 \mathbf{Z}) \\ \operatorname{Bexp}(-\mathbf{i} 2\mathbf{q}_0 \mathbf{Z}) & \mathbf{\varepsilon} \end{bmatrix} \begin{bmatrix} \mathbf{E}_1 \\ \mathbf{E}_2 \end{bmatrix}$$

$$\dots (26)$$

Here it is not possible to remove the space dependence by assuming a variation of the form e^{ikz} for E since \tilde{c} also depends on Z. However, using a trial solution of the type $(e^{ikz}, 0)$ one can show that the effect of the dependence of \tilde{c} on Z is to convert a wave of the '1' polarisation into '2' with a shift of wave vector down by $2q_0$. Similarly using a trial solution of the type $(0, e^{ikz})$ one finds that it converts a '2' wave into '1' wave with an upward shift of the wave vector by $2q_0$. Thus it is obvious that a superposition of the type

is **closed in** the sense **that** each of the waves **appearing** in it is converted into the other, and can therefore **satisfy** (26) with proper **choice** of a and b. The mixing of the two wave vectors differing by $2q_0$ is a consequence of Bragg reflection. Substituting (27) into (26) gives

$$\begin{pmatrix} (k + q_0)^2 - \frac{\varepsilon w^2}{c^2} & -B \frac{w^2}{c^2} \\ & & & \\ -B \frac{w^2}{c^2} & (k - q_0)^2 - \frac{\varepsilon w^2}{c^2} \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix} = 0 \quad (28)$$

Here the condition to find We ratio **a/b** is given **by**

$$[(k + q_0)^2 - K_m^2][(k - q_0)^2 - K_m^2] - B^2 K^4 = 0$$
 (29)

where K = w/c is the wave vector in free space and $K_m = E^{\frac{1}{2}}w/c$ is the wave vector corresponding to frequency w in a medium of dielectric constant ε . (29) is a quadratic equation in k^2 having roots

$$k_{2}, k_{1} = [K_{m}^{2} + q_{0}^{2} \pm (4K_{m}^{2}q_{0}^{2} + B^{2}K^{4})^{\frac{1}{2}}]^{\frac{1}{2}}$$
 (30)

The value of **a/b corresponding** to each of these can be obtained **from** (28)

$$a/b = \frac{B K^2}{(k + q_0)^2 - K_m^2} = \frac{(k - q_0)^2 - K_m^2}{B K^2}$$
 (31)

Equations (30) and (31) completely determine the exact solutions of equation (23).

The solutions are interpreted physically in the following way:

When B = 0, then the wave vectors of '1' wave and '2' wave reduces to K_m and hence the two circular waves travel with the same velooity in the medium. When $B \neq 0$, from (30), $k_{1,2}$ and $K_m \neq q_0$ differ by quantities of the order of B^2 . From (31), one gets for the k_2 solution $a/b \sim B^2/B \sim B$ and for k_1 solution, $b/a \sim B$. Thus they are no longer pure oiraular waves, still one oan continue to refer the k_1 solution as '1' dominant (u_1) and k_2 solution a^5 '2' dominant (u_2) . Each normal wave consists of a combination of the two circularly polariaed light with one dominating over the other and differing in wave vector by $2q_0$.

Therefore equations (27), (30) and (31) can

be written aa

$$u_{1} = [\exp(iK_{1}Z), d \exp\{i(K_{1} - 2q_{0})Z\}]$$

$$u_{2} = [f \exp\{i(K_{2} + 2q_{0})Z\}, \exp(iK_{2}Z)] \qquad (32)$$

where the wave vectors of the dominant components are

$$K_{1} = k_{1} + q_{0} = q_{0} + [K_{m}^{2} + q_{0}^{2} - (4K_{m}^{2}q_{0}^{2} + B^{2}K^{4})^{\frac{1}{2}}]^{\frac{1}{2}}$$

$$K_{2} = k_{2} - q_{0} = -q_{0} + [K_{m}^{2} + q_{0}^{2} + (4K_{m}^{2}q_{0}^{2} + B^{2}K^{4})^{\frac{1}{2}}]^{\frac{1}{2}}$$
... (33)

and

$$d = \frac{K_1^2 - K_m^2}{B K^2}, \quad f = \frac{K_2^2 - K_m^2}{B K^2} \quad (34)$$

The dominantly **right circular** and dominantly left **circular waves** travel with different wave **vectors** and result in **optical** rotation.

The optical rotation per unit length is

$$\int = \frac{K_1 - K_2}{2} = \frac{1}{2} \left[\left\{ K_m^2 + q_0^2 - (4K_m^2 q_0^2 + B^2 K^4)^{\frac{1}{2}} \right\}^{\frac{1}{2}} - \left\{ K_m^2 + q_0^2 + (4K_m^2 q_0^2 + B^2 K^4)^{\frac{1}{2}} \right\}^{\frac{1}{2}} + 2q_0 \right]$$

$$\dots (35)$$

The expressions (32), (33), (34) and (35) are exact and explain total reflection as well as anomalous rotatory dispersion in a spiral dielectric medium of semi-infinite thickness. The rigorous electromagnetic theory has been extended to finite thickness by Nityananda and Kini (1973). For finite thickness the theory leads to complicated oet of formulae and it is very difficult to extract measurable parameters.

4. Comparison of the dynamical theory with the electromagnetic theory

To compare the dynamical theory with the exact theory quantitatively, one requires calculations of the valuers of important **optical** parameters, **viz.**, (a) reflection coefficient R, (b) the wave vectors K_{R} and K_{L} , and (c) rotatory

99 19

power *e* as functions of wavelength both for samples of finite and semi-infinite thioknese. The parameters chosen for the calculation are $\mu = 1.5$, $\Delta \mu = 0.07$ and $P = 0.3333 \mu m$. The calculations were made on an IBM-360 computer using Fortran IV language. Figure 2 shows R as a function of wavelength. The semi-infinite sample gives the well.-known flat topped curve of the dynamical theory, while the thin sample gives a principal maximum accompanied by subsidiary fringes, which have been observed experimentally (Dreher, Meier and Saupe 1971, Chandrasekhar and Prasad 1971). More recently, on the basis of dynamical theory, Mazkedian et al. (1976) have reinterpreted these fringes in analogy with 'Pendellösung fringes¹ that occur in X-ray refleation by perfect crystals. Figure 2 also presents the values computed from the exact theory of Nityananda. In the exact theory, the external isotropic medium (external to the cholesteric specimen) is assumed to have a refractive index of 1.5, so that the contribution of the ordinary Fresnel reflection coefficient at the cholesteric/isotropic interface is eliminated. Figure 3 gives the wave vectors K_R and K_L (i.e.,

Figure 2: Reflection coefficient R versus wavelength À in the non-absorbing case: (a) semi-infinite medium, (b) film of thickness 25 P. Curves are derived from the dynamical theory; circles represent values computed from the exact theory.





<u>Figure 3:</u> The wave vectors $\mathbf{K}_{\mathbf{R}}$ and $\mathbf{K}_{\mathbf{L}}$ of the normal waves as functions of λ in a cemi-infinite non-absorbing medium. Curves are derived from the dynamical theory; circles represent values computed from the exact theory.

phase retardation per pitch) of the normal waves as functions of wavelength in the semi-infinite medium. It also gives the values computed from the exact theory. In Figure 4, the optical rotatory power ? in the semi-infinite and finite samples calculated from the dynamical theory and exact theory are presented. (I am grateful to R. Nityananda and UD. Kini for providing numerical resulte computed from the exact theory.) As in the long pitch regime (see Chapter I, Figure 3), the rotatory power is again a function of the thickness of the sample. This has been confirmed experimentally (Martin & Cano 1974) (see Figure 5).

One can see that the results obtained from the dynamical model are in conformity with those from the detailed electromagnetic theory. However, the simple dynamical approach presented here has certain limitations viz., (1) it is developed for small c and therefore does not hold good for wavelengths far away from the reflection band, (2) it is strictly valid for integral values of the pitch, (3) it fails when the film thickness is very small (or when the extinction length is of the order of pitch) as the assumption that the normal waves are circularly polarised is than no longer justified. These limitations can be removed by including the effect of multiple reflections within the n layers per turn of the

.



Figure 4: Rotatory power \mathcal{G} versus λ in the non-absorbing case; (a) semi-infinite medium, (b) film of thickness 25 P. Curves are derived from the dynamical theory; circles represent values computed from the exact theory.



Figure St Optical rotatory dispersion in cholesteryl benzoate containing 50 per cent by weight p-azoxyanisole for various sample thicknesses: (I) 1.7 µm, (II) 4 µm and (III) 7 µm. (Martin and Cano 1974)

helix, which has been neglected in the discussion. The simple difference equations then become matrix difference equations and the resulting solutions can be shown to be fully equivalent to those of the rigorous treatment. However, the calculations presented earlier indicate that this more elaborate formulation of the theory is probably not necessary for most practical problems.

References

- Aihare, M. and Inaba, H. 1971 Optics Commun. 3, 77.
- Chandrasekhar, S. and Srinivasa Rao, K.N. 1968 Acta Cryst., 445.
- Chandrasekhar, S. and Shashidhara Prasad, J. 1971 Mol. Cryst. Liquid Cryst., 115.
- Chandrasekhar, S. and Ranganath, G.S. 1974 Mol. Cryst. Liquid Cryst. 25, 195.
- Chandrasekhar, S., Ranganath, G.S. and Suresh, K.A. 1973 Proceedings of the International Liquid Crystals Conference, Bangalore - Pramana Suppl. 1, 341.
- Darwin, C.G. 1914 Phil. Mag. 27, 315, 675; 1922 <u>1b1d</u> 43, 800.
- de Vries, H. 1951 Aota Cryst. 4, 219.
- Dreher, R., Meier, G. and Saupe, A. 1971 Mol. Cryst. Liquid Gryst., 17.
- Joly, G. 1972 'Contribution a' l'etude de la propagation de certaines ondes electromagnetiques dans lea piles de reusch', Thesis, Univ. of Soienoe and Technology, Lille, France.
- Kats, E.I. 1971 Sov. Phys.-JETP, 32, 1004.

Marathay, A.S. 1971 J. Opt. Soc. Amer. <u>61</u>, 1363.

Martin, J. and Cano, R. 1974 C.R.Acad.Sci. 278B, 219.

- Mauguin, M.C. 1911 Bull. Soc.Franc. Miner. Crist. 34, 71.
- Mazkedian, S., Melone, S. and Rustichelli, F. 1976 Phys. Rev. <u>A14</u>, 1190.

These authors have aleo worked out the effects of pitch gradient on dynamical reflection -J. de Physique, 36, C1-283 (1975).

- Nityananda, R. 1973 Mol. Cryst. Liquid Cryst. 21, 315.
- Nityananda, R. and Kini, U.D. 1973 Proceedings of the International Liquid Crystals Conference, Bangalore - Pramana Supplement <u>1</u>, 311.
- Oseen, C.W. 1933 Trans. Faraday Sou. 29, 833.