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Theory of Rotatory Dispersion of Cholesteric Liquid Crystals†

S. CHANDRASEKHAR and J. SHASHIDHARA PRASAD

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Abstract—A theory is developed of the circular dichroism and rotatory power of thin films of cholesteric liquid crystals. It is an extension of the treatment for infinitely thick specimens discussed in a previous paper. The theory predicts that the circular dichroism of a thin film plotted as a function of wavelength should exhibit a principal maximum accompanied by subsidiary maxima, and that the rotatory dispersion should be anomalous. Experimental circular dichroism and rotatory dispersion curves are presented for cholesteric cinnamate, cholesteryl-2-propyn-1-yl carbonate and spectratherm. The results are in qualitative agreement with the theory both inside and outside the region of reflexion. The rotatory dispersion curves are also in accord with the de Vries equation outside the region of reflexion.

1. Introduction

The first theory to be proposed of the very high rotatory power of cholesteric liquid crystals and its relation to the reflexion of circularly polarized light was by de Vries.⁽¹⁾ According to this theory, the spectral width of total reflexion of circularly polarized light incident along the optic axis of an infinitely thick specimen is $P\Delta\mu$, and the rotatory power outside the region of reflexion is

$$\rho = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2[1-(\lambda/\lambda_0)^2]}, \tag{1}$$

where P is the pitch of the helical structure, $\Delta\mu$ the layer birefringence, λ the wavelength in vacuum and $\lambda_0 = \mu P$, μ being the mean refractive index. The predictions of the theory are in qualitative agreement with observations. When $\lambda \ll P$, (1) reduces to

$$\rho = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2}, \tag{2}$$

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a relation first derived by Mauguin,(2) which has been verified quantitatively by Robinson(3) and by Cano and Chatelain.(4,5)

Another somewhat simpler and more direct explanation of the reflexion and rotatory power was put forward by us(6) making use of a modified form of Darwin's dynamical theory of X-ray diffraction.(7,8) This theory leads to a spectral width of total reflexion from an infinitely thick specimen equal to $Q\lambda/\pi$ and the following formulae for the rotatory dispersion:

outside the region of reflexion ($\lambda < \lambda_0 - Q\lambda/2\pi$ or $\lambda > \lambda_0 + Q\lambda/2\pi$),

$$\rho = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2} + \frac{\pi(\lambda-\lambda_0)}{P\lambda}\left[1-\left(1-\frac{Q^2}{\epsilon^2}\right)^{1/2}\right]; \tag{3}$$

inside the region of reflexion ($\lambda_0 - Q\lambda/2\pi < \lambda < \lambda_0 + Q\lambda/2\pi$),

$$\rho = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2} + \frac{\pi(\lambda-\lambda_0)}{P\lambda}, \tag{4}$$

where $\epsilon = -2\pi(\lambda-\lambda_0)/\lambda$ and Q is the reflexion coefficient per pitch of the cholesteric structure.

Strictly, the rotatory power cannot be measured for an infinitely thick specimen inside the reflexion band since one of the circular components is completely attenuated. Nevertheless, (4) is theoretically significant in that it shows how the phase difference between the two opposite circular waves (or, equivalently, the azimuth of the major axis of the elliptically polarized wave) varies with wavelength within the reflexion band. Far away from the region of reflexion on either side, the second term in (3) becomes small and the rotatory power approaches the normal value given by (2).

In discussing the form of the rotatory dispersion curve in the previous paper,(6) the dependence of Q and $\Delta\mu$ on each other and on λ were neglected. In the present paper, we shall show that inclusion of these factors modifies the rotatory dispersion curve slightly so that it now bears a close resemblance to that derived from the de Vries equation outside the reflexion band. We shall also extend the theory to films of finite thickness and compare the results with detailed observations on some cholesteric materials.

2. Theory of Reflexion

We assume the cholesteric structure to be built up of a large number of thin birefringent layers with the principal axes of the successive layers turned through a small angle β. By the application of the Jones calculus, it was shown$^{(6)}$ that when reflexion is negligible the optical rotatory power is given by (2), the negative sign indicating that the sign of the rotation is opposite to that of the helical twist of the structure.

Let us suppose that the structure is right handed. Right circular light incident along the optic axis of such a structure is reflected without change of sense of circular polarization when $\lambda_0 = \mu_d P$, where μ_d is the refractive index for right circular light. Left circular light, on the other hand, is not reflected.

The reflexion coefficient at the boundary between the vth and $(v+1)$th layers is

$$|q| = \frac{\mu'_{v+1} - \mu'_v}{\mu'_{v+1} + \mu'_v}, \tag{5}$$

where μ'_{v+1} and μ'_v are the refractive indices of the $(v+1)$th and vth layers for a given direction of the electric vector. If we assume that $\mu'_{v+1} - \mu'_v \simeq \beta \Delta\mu$,

$$|q| \simeq \frac{\beta \Delta\mu}{2\mu},$$

where $\mu = \frac{1}{2}(\mu_1 + \mu_2)$, $\Delta\mu = \mu_1 - \mu_2$, μ_1 and μ_2 being the principal refractive indices of a layer. The reflexion coefficient per thickness P of the liquid crystal is then

$$Q_0 = n\,|q| \simeq \frac{\pi \Delta\mu}{\mu}, \tag{6}$$

since $n\beta = 2\pi$, where n is the number of molecular layers per pitch of the helix.

The general expression for the variation of the refractive index with direction is

$$\frac{1}{\mu'^2} = \frac{\cos^2\alpha}{\mu_1^2} + \frac{\sin^2\alpha}{\mu_2^2}, \tag{7}$$

which for the present calculation may be represented quite well by the relation$^{(9)}$

$$\mu'_v = \mu - \tfrac{1}{2}\Delta\mu \sin 2v\beta. \tag{8}$$

Substituting (8) in (7) and averaging over the different layers,

$$|q| \simeq \frac{\beta \Delta\mu}{\pi\mu}$$

and

$$Q_0 \simeq \frac{2\Delta\mu}{\mu}, \tag{9}$$

which is a slightly better approximation than (6).

The effect of multiple reflexions may be calculated by setting up difference equations similar to those formulated by Darwin.(7,8) Such a procedure is valid because circularly polarized light is propagated along the optic axis without change of form, and the interference of multiply reflected waves with one another and with the primary wave can be evaluated directly. For the convenience of development of this theory, we shall regard the liquid crystal as consisting of a set of parallel planes spaced P apart and ascribe a reflexion coefficient $-iQ$ to each plane for right circularly polarized light at normal incidence. (The relation between Q and Q_0 will be discussed presently.) Let T_r and S_r be the complex amplitudes of the primary and reflected waves at a point just above the rth plane, the topmost plane being designated by the serial number zero. Neglecting the normal absorption coefficient, which is in fact extremely small in the visible spectrum for most of these compounds,(10) the difference equations may then be written as

$$S_r = -iQT_r + \exp(-i\phi)S_{r+1}, \tag{10}$$

$$T_{r+1} = \exp(-i\phi)T_r - iQ\exp(-2i\phi)S_{r+1}, \tag{11}$$

where $\phi = 2\pi\mu_d P/\lambda$. The reflexion coefficient is here taken to be the same on both sides of the plane. Replacing r by $(r-1)$ in (10) and (11), substituting and simplifying, we obtain

$$T_{r+1} + T_{r-1} = yT_r \tag{12}$$

$$S_{r+1} + S_{r-1} = yS_r \tag{13}$$

where $y = \exp(i\phi) + \exp(-i\phi) + Q^2\exp(-i\phi)$.

Suppose that the film consists of m planes. Putting $S_m = 0$, we have from (13)

$$S_{m-2} = yS_{m-1},$$

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

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

and $$S_0 = \left[y^{m-1} - \frac{(m-2)}{1!} y^{m-3} + \frac{(m-4)(m-3)}{2!} y^{m-5} - \ldots \right] S_{m-1}$$

$$= f_m(y) S_{m-1} \quad \text{(say)}. \tag{14}$$

Similarly, from (11) and (12),

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

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

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

and $$T_0 = [f_m(y) \exp(i\phi) - f_{m-1}(y)] T_m. \tag{15}$$

Since from (10),

$$S_{m-1} = -iQ T_{m-1} = -iQ \exp(i\phi) T_m,$$

the ratio of the reflected to the incident amplitudes is

$$\frac{S_0}{T_0} = - \frac{iQ f_m(y) \exp(i\phi)}{f_m(y) \exp(i\phi) - f_{m-1}(y)}. \tag{16}$$

Let us assume a relation in the form $T_{r+1} = x T_r$, so that x satisfies

$$x + \frac{1}{x} = y = \exp(i\phi) + \exp(-i\phi) + Q^2 \exp(-i\phi). \tag{17}$$

We have seen that the reflexion condition is $\mu_d P = \lambda_0$ or $\phi_0 = 2\pi$. Accordingly we may write

$$\phi = 2\pi\lambda_0/\lambda = \phi_0 + \epsilon, \tag{18}$$

where $$\epsilon = -2\pi(\lambda - \lambda_0)/\lambda, \tag{19}$$

which is a small quantity in the neighbourhood of the reflexion. Therefore, from (17)

$$x + \frac{1}{x} = \exp(i\epsilon) + \exp(-i\epsilon) + Q^2 \exp(-i\epsilon). \tag{20}$$

This suggests that in the neighbourhood of the reflexion we may put

$$x = \exp(-\xi) \exp(-i\phi_0) = \exp(-\xi), \tag{21}$$

where ξ is small and may be complex. From (20) and (21),

$$\xi \simeq \pm (Q^2 - \epsilon^2)^{1/2}. \tag{22}$$

When

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

the series in (14) is summable (see, e.g., Ref. 11) and is given by

$$f_m(y) = \frac{\sinh m\xi}{\sinh\xi}. \tag{23}$$

Substituting in (16) and simplifying

$$\frac{S_0}{T_0} \simeq \frac{-iQ\exp(i\epsilon)}{i\epsilon + \xi\coth m\xi}, \tag{24}$$

or

$$R = \left|\frac{S_0}{T_0}\right|^2 = \frac{Q^2}{\epsilon^2 + \xi^2\coth^2 m\xi}. \tag{25}$$

For an infinitely thick specimen,(6)

$$\frac{S_0}{T_0} = -\frac{Q}{\epsilon \pm i\xi} = \frac{Q}{\epsilon \pm i(Q^2 - \epsilon^2)^{1/2}} \tag{26}$$

When $-Q < \epsilon < Q$, ξ is real and

$$R_{\text{inf}} = |S_0/T_0|^2 = 1.$$

The reflexion is therefore total within this range. The spectral width $\Delta\lambda$ of total reflexion is $Q\lambda/\pi \simeq Q_0\lambda_0/\pi$. If we use (6), $\Delta\lambda = P\Delta\mu$, in exact agreement with the de Vries theory. The slightly better approximation (9) gives $\Delta\lambda = 2P\Delta\mu/\pi$. Outside this range, ξ is imaginary and the reflexion falls off rapidly on either side. When $\lambda > \lambda_0$, ϵ is negative and hence the negative value of the square root in the denominator of (26) has to be taken because R_{inf} can never exceed unity; when $\lambda < \lambda_0$, the positive root has to be taken.

So far, we have regarded the liquid crystal as consisting of a set of parallel planes, each with a reflexion coefficient $-iQ$. Clearly $-iQ$ represents the over-all effect of the reflexions from n molecular layers and is related to $-iq$ the reflexion coefficient of a single molecular layer as follows:

$$\begin{aligned} Q &= q[1 + \exp(-2i\delta) + \exp(-4i\delta) + \cdots \exp(-2ni\delta)] \\ &= \frac{q[1 - \exp(-2ni\delta)]}{[1 - \exp(-2i\delta)]}, \end{aligned}$$

where $\delta = 2\pi\mu_a P/n\lambda = \emptyset/n$. Therefore

$$|Q|^2 = \frac{q^2\sin^2 n\delta}{\sin^2\delta} = n^2q^2\left(\frac{\sin\emptyset}{\emptyset}\right)^2.$$

In the neighbourhood of the reflexion,

$$Q^2 = Q_0^2\left(\frac{\sin\epsilon}{\epsilon}\right)^2. \tag{27}$$

Using (25), (26) and (27), theoretical reflexion curves have been drawn with $Q_0 = 0.1$ (or $\Delta\mu = 0.075$) and $\lambda_0 = 6000 \times 10^{-8}$ cm for various values of m; these are shown in Fig. 1. With increasing film thickness the peak intensity of the principal maximum increases while its width decreases; at the same time the subsidiary maxima increase in number and get closer together, until for the very thick film the principal and subsidiary maxima merge to give a single flat-topped maximum. The width of total reflexion for the very thick specimen is $2P\Delta\mu/\pi$.

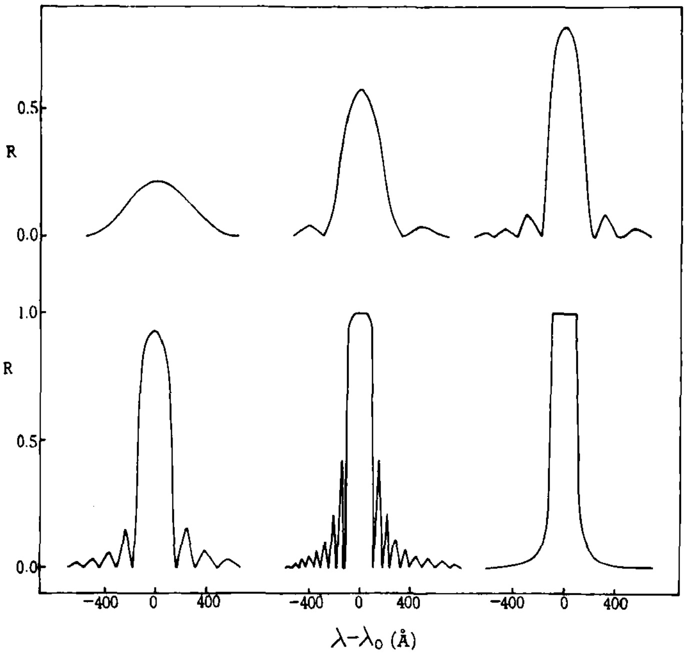

Figure 1. Theoretical reflexion curves for films of thickness mP; $m = 5, 10, 15$ (top row), 20, 40 and ∞ (2nd row).

3. Circular Dichroism

From (15) and (23),

$$\frac{T_m}{T_0} = \left[\exp(i\epsilon)\frac{\sinh m\xi}{\sinh \xi} - \frac{\sinh (m-1)\xi}{\sinh \xi}\right]^{-1}$$

$$\simeq \frac{\xi \operatorname{cosech} m\xi}{i\epsilon + \xi \coth m\xi}. \tag{28}$$

It is easily verified that

$$\left|\frac{T_m}{T_0}\right|^2 + \left|\frac{S_0}{T_0}\right|^2 = 1.$$

The circular dichroism is therefore

$$D = \frac{R}{2-R}. \tag{29}$$

D plotted as a function of wavelength should exhibit primary and secondary maxima similar to the reflexion curve *except that the peaks will be less pronounced.*

4. Anomalous Rotatory Dispersion

In the region of normal dispersion, the optical rotation per thickness P of the liquid crystal is $\frac{1}{2}(\phi_d - \phi_l)$ and the rotatory power in radians per cm

$$\rho = \frac{1}{2P}(\phi_d - \phi_l) = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2},$$

where ϕ_d, ϕ_l are the phase retardations for right and left circular light. Here a clockwise rotation as seen along the direction of propagation of light is taken as positive.

Near the region of reflexion, the dynamical theory predicts that the right circular component suffers anomalous phase retardation. Left circular light, on the other hand, exhibits normal behavior throughout and, as a consequence, the rotatory dispersion is anomalous in the vicinity of the reflexion.

The phase of the right circular wave can be evaluated from (28). The ratio of the transmitted to the incident amplitudes may be expressed as

$$\frac{T_m}{T_0} = A \exp\{-im(\phi_0 + \psi)\} = A \exp(-im\,\psi),$$

where $$\tan m\,\psi = \frac{\epsilon}{\xi \coth m\xi}.$$

The optical rotation per thickness P is

$$\tfrac{1}{2}(\phi_0 + \psi - \phi_l) = \tfrac{1}{2}[(\phi_d - \phi_l) + (\psi - \epsilon)],$$

from (18). Hence the rotatory power in radians per cm

$$\rho = -\frac{\pi(\Delta\mu)^2 P}{4\lambda^2} + \frac{(\psi - \epsilon)}{2P}. \tag{30}$$

Far away from the region of reflexion *on either side*, i.e., when $\epsilon^2 \gg (Q_0^2 \sin^2 \epsilon/\epsilon^2)$, $\xi \simeq i\epsilon$ from (22), $\psi \simeq \epsilon$ and the rotatory power reduces to the normal value.

The theoretical rotatory dispersion curve is illustrated in Fig. 2 for $m = 10$, $\lambda_0 = 6000 \times 10^{-8}$ cm, $P = 4000 \times 10^{-8}$ cm (or $\mu_d = 1.5$), $Q_0 = 0.1$ (or $\Delta\mu = 0.075$ at λ_0). The layer birefringence $\Delta\mu$ was assumed to vary linearly from 0.1 at λ4000 Å to 0.075 at λ6000 Å and extrapolated to longer wavelengths. The curve gives the rotatory power (in degrees/mm) of a right-handed structure using the standard sign convention according to which a clockwise rotation as seen by an observer looking at the source of light is taken as positive.

The corresponding curve for the very thick specimen given by (3) and (4) is shown as a broken curve in Fig. 2. As mentioned earlier, the curves drawn in the previous paper⁽⁶⁾ had neglected the relation between Q_0 and $\Delta\mu$; Q_0 was taken as 0.05 and $\Delta\mu$ as 0.15, whereas according to (9) $\Delta\mu$ should have been 0.0375 at λ_0. Moreover, the variation of $\Delta\mu$ with λ had been ignored. Consequently, the shape of the curve was somewhat distorted and the theory seemed to show a zero of rotatory power close to the reflexion band on the long wavelength side. In fact, when these factors are properly taken into account, the curve tends to be almost asymptotic with the zero rotatory power line as can be seen in Fig. 2. It is interesting to note that the curve is now closely similar to what is expected from the de Vries equation (1).

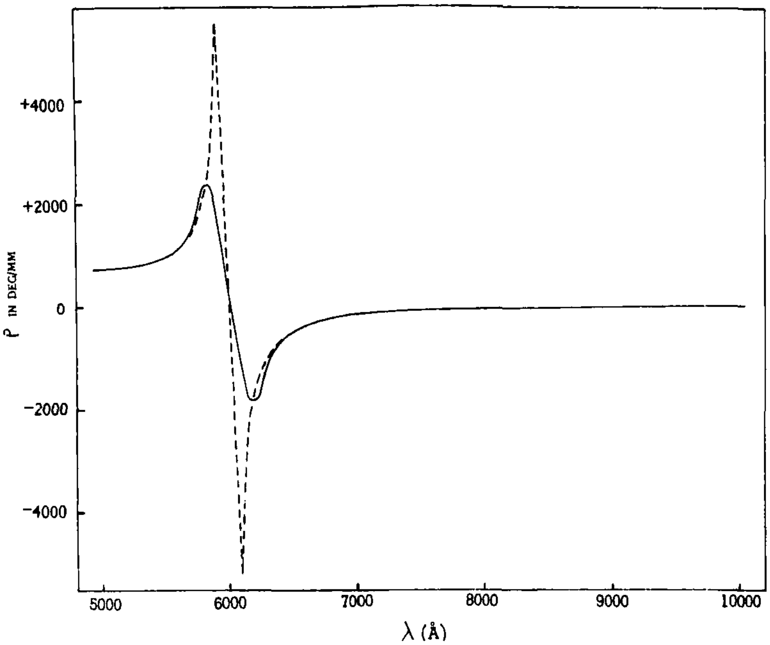

Figure 2. Theoretical rotatory dispersion curves; $m = 10$ (full curve), $m = \infty$ (broken curve).

5. Comparison with Experiment

Circular dichroism and anomalous rotatory dispersion curves right through the reflexion band have been reported by Mathieu(12) for cholesteric cinnamate and by Fergason(10) for a mixture of cholesteryl benzoate, acetate and palmitate. Their data are in broad agreement with the predictions of our theory both inside and outside the reflexion band.

We have carried out somewhat more detailed measurements on cholesteric cinnamate, cholesteryl-2-propyn-1-yl carbonate (CPC) and a sample of spectratherm supplied by Westinghouse Electric Corporation, Pittsburgh, Penna. (When the spectratherm was supplied five years ago it had a temperature response of 20°–60 °C from red to violet, but in course of time the temperature range changed gradually. At present the reflexion occurs at about 4500 Å at room temperature. The composition of the sample is not known.) The measurements were made on 'plane texture' preparations[(13)] between microscope slides. No spacers were used. The polarimetric arrangement consisted of a Perkin-Elmer-Hitachi (model 139) monochromator, linear polarizer and analyzer, and a sensitive phototransistor detector. The relative rotations could be determined to ± 4′ of arc from 4000–10000 Å. Small areas of the film, about 2×2 mm^2, were used for the experiments. That the areas were optically homogeneous were checked by the uniformity and perfection of the extinction for wavelengths outside the reflecting region. The film thickness was measured by forming interference fringes between the glass surfaces in air gaps. Since no spacers were used, the thickness measurements are not expected to be accurate to better than ± 15%.

The circular dichroism was obtained by measuring the intensity of the transmitted light for right and left circular polarizations and applying the formula

$$D = \frac{I_l - I_r}{I_l + I_r}.$$

This procedure eliminates errors arising from reflexions from glass surfaces, spectral variations of the source and of the sensitivity of the detector, etc. However it suffers from the limitation that the quarter wave plate has to be achromatic for the measurements to be quantitatively precise. Since D involves the difference of two intensity measurements, which is quite small for the subsidiary maxima, the error in the heights of the maxima may be expected to be appreciable. Moreover this error will vary with wavelength. No correction has been applied for this in our measurements and thus the circular dichroism curves presented here are intended for qualitative comparison with the theory.

Observations were made on a number of specimens of cholesteric cinnamate, CPC and spectratherm. Typical results are shown in Figs. 3–5, and as can be seen they reproduce the features predicted by theory. The rotatory dispersion curves are also in qualitative agreement with the de Vries equation outside the region of reflexion.

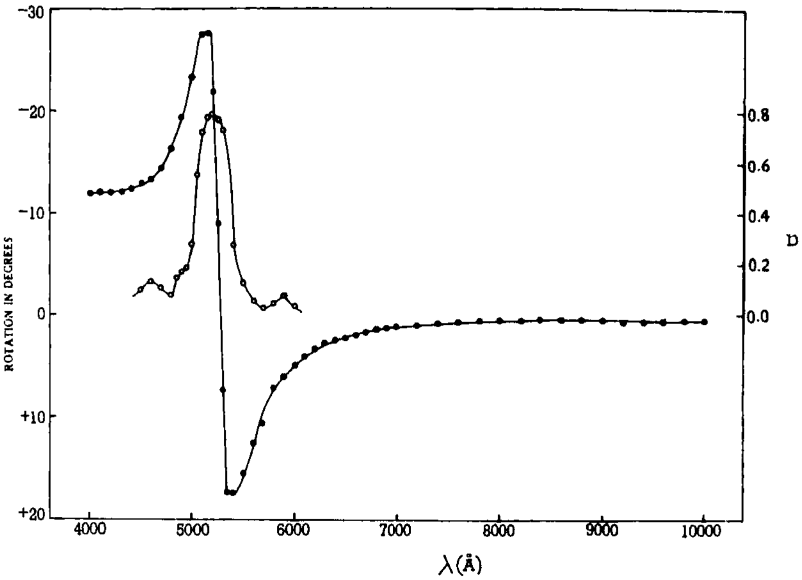

Figure 3. Circular dichroism and rotatory dispersion of cholesteric cinnamate at 177 °C. Thickness ~3μ.

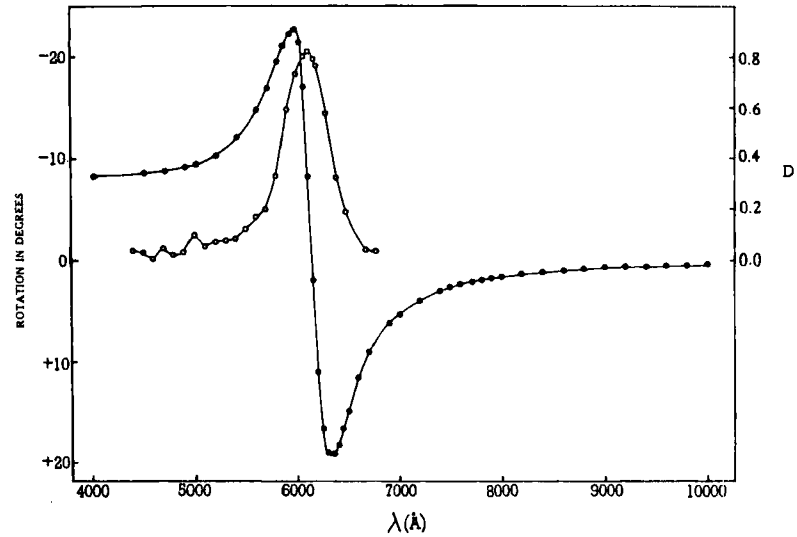

Figure 4. Circular dichroism and rotatory dispersion of cholesteryl-2-propyn-1-yl carbonate at room temperature (supercooled). Thickness ~4 μ.

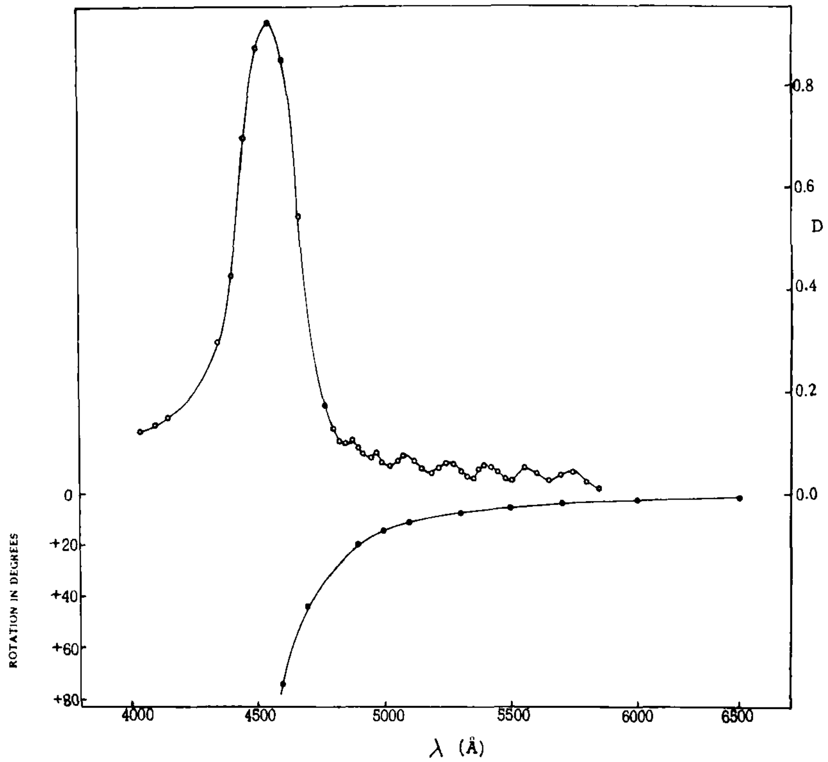

Figure 5. Circular dichroism and rotatory dispersion of spectratherm at room temperature. Thickness ~18 μ.

6. Concluding Remarks

Since the derivation of the Mauguin formula (either by the use of the Poincare sphere(2) or the Jones calculus(6)) would appear to be valid irrespective of whether λ is greater than or less than P, the rotatory power may be expected to reduce to the normal value when reflexion is negligible. Thus, in addition to the reversal of sign of rotation inside the reflexion band, there should be a second reversal of sign on the long wavelength side of the band. Such a behavior is not predicted by the de Vries equation in its present form. However, since $(\Delta\mu/\lambda)^2$ decreases very rapidly with increase of λ, the curve tends to be nearly asymptotic with the zero rotatory power line (Fig. 2). This was indeed found to be the case experimentally up to

10000 Å for all the specimens examined. Nevertheless, in principle, a change of sign of rotation should be observable under favourable circumstances. Neville and Caveney⁽¹⁴⁾ have recently reported such a behavior in the rotatory dispersion curves of some beetle exocuticles which form helical structures similar to cholesteric liquid crystals.† It would be of interest to confirm these observations in other liquid crystalline systems.

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† These authors have wrongly stated that there is a numerical error in our formulae for the rotatory power.⁽⁶⁾ The confusion has arisen because they have used different units for P and λ.

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