

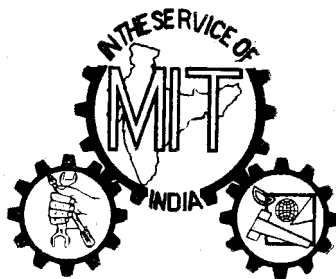
STUDY OF LIQUID CRYSTALS

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Thesis Report submitted in partial fulfilment of the requirements for the award of M. Sc. (Engg.) Degree in Electrical and Electronics Engineering (Applied Electronics) of the University of Madras.

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This is to certify **that** Sri G. Vasudevan
carried out some studies in our Laboratory on
the optical properties of cholesteric liquid
crystals as a part of the experimental work
required for his dissertation for the M.Sc.
Engineering Degree (Madras University).

S. Chandrasekhar

Dedicated to

my Uncle, Shri. P. Balasubramaniam,

ACKNOWLEDGMENT

" Prof. S. Chandrasekhar !

**A hundred lives would not be sufficient to pay my
deep debt of gratitude to you
..... If the Indian Ocean were an inkstand, the
highest mountain of the Himalayas the pen, the
earth the Scroll, and time itself the writer,
still it will not express my gratitude to you."**

Swami Vivekananda.

**This work was done at the Raman Research Institute,
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P R E F A C E

The discovery of the cholesteric type of liquid crystal coincides with the discovery of the liquid crystalline phase itself by Reintitser in 1888, for the first few compounds were esters of cholesterol.

The cholesteric phase has a structure in which the molecules are arranged in layers each layer consisting of molecules aligned approximately parallel to one another. However, the direction of alignment of the molecules rotates continuously on moving along with the normal to the layers (fig.). A homogeneous specimen (referred to as 'Plane texture' or 'structure a plans' of this phase) exhibits certain unique optical properties that readily distinguish it from the other two liquid crystalline phases. In this classical memoir on liquid crystal Friedel described the properties of the plane structure in the following words (1922).

'La structure a plans des corps cholesteriques possede des caracteres tres particuliers, en partie remarques depuis long temp, et qui font de la phase cholesterique un type de matiere bien singulier. Elle comporte, en premier lieu, des plages uniformes, qui dans la structure a plans parfaitement reguliere existent seules et sont orientees normalement a l'axe optique,

ces plages, conservées en lumière naturelle, par réflexion diffusent de vives couleurs variables selon l'incidence. Entre autres, elles montrent de des couleurs qui ne changent pas par rotation de la platine et qui sont dues en partie à un pouvoir rotatoire en général excessivement fort, et en partie aussi, comme nous verrons, à une autre cause. Bien régulières, elles se montrent, dans des conditions convenables d'observation, composées de strates d'égales épaisseurs limitées par des surfaces de discontinuité (plans de grandjean) ces trois caractères, diffusion de couleurs, pouvoir rotatoire et présence des plans de grandjean sont intimement liés l'un à l'autre et n'existent pas l'un sans l'autre."

Since its discovery there have been a number of experimental investigations and a few theoretical studies on the remarkable optical properties of this phase, but it would be fair to say that many aspects of these phenomena have not yet been fully understood. A brief summary of the salient results of these investigation is given below.

In Chapter (1) first review the experimental facts regarding the optical properties of cholesteric liquid-crystal. viz., the very high rotatory power, the selective reflection of circularly polarized light at normal incident and the anomalous rotatory dispersion around the

region of reflection. We then discuss the previous theoretical work of Mauguin (1911-1913) and de Vries (1951) Mauguin calculated the rotatory power of a twisted nematic liquid crystal (produced by rotating the coverslip about an axis normal to the preferred direction of the molecules) by assuming a model of a helically twisted birefringent medium and applying the Poincare sphere and rolling cone method. Mauguin's work provided the first clue to the origin of the very high rotatory power of the cholesteric phase. The experimental work by Robinson and Ward (1956, 57, 58, 61) and Chatelain and Cano (1964, 61) showed that the Mauguin's formula is valid for the cholesteric liquid crystal where P , where P is the pitch of the helix, but it does not hold good near the region of reflection when becomes comparable to P . In 1951 de Vries proposed a theory of light propagation along the optic axis of cholesteric structure assuming a model in which the principal dielectric constant twist continuously in the form of a helix. The theory predicted total reflection of circularly polarized light from thick specimens over a spectral range of few hundred Å and the reversal of sign of the rotatory power on opposite sides of the reflection band. These predictions are in broad agreement with the observed facts.

In chapter II we discuss a new approach put forward by Chandrasekhar and Srinivasa Rao (1968) for explaining the optical properties of cholesteric liquid crystals for light propagation along with optic axis for infinitely thick specimen. The effect of a plate of anisotropic material such as a crystal, on a collimated beam of polarised light may be represented mathematically as a linear transformation of the components of the electric vector of the light. The effect of a retardation plate, or an anisotropic absorber, or of a crystal possessing optical activity may be represented as a matrix operator which operates on the electric vector of the incident light. A plane wave of light characterised by the phases and amplitudes of the two by two matrix, with matrix elements in general complex. The above ideas were first developed in detail with various applications by R.C. Jones in a series of papers (1941 - 56) and the mathematical method has come to be known as the Jones calculus. We shall now develop a theory of the optical properties of an infinitely thick cholesteric liquid crystal for light propagation along the optic axis by the application of the Jones Calculus.

In Chapter III we shall discuss the extension of the above theory to films of finite thickness, as encountered in actual practice, as given by Chandrasekar

and Shashidhara Prasad (1970). The optical properties of thin films may be expected to be slightly different from that of very thick specimen. We shall therefore *discuss* ^{the extension} ~~extend~~ the theory of reflection, circular dichroism and rotatory dispersion to take into account the finite thickness of the film.

In chapter iv we shall give some detailed experimental data for rotatory power and reflection. As we shall see, the theoretical predictions are in agreement with the observed phenomena.

In Chapter v we discuss the theory of light propagation normal to the optic axis of a cholesteric liquid crystal. We have so far investigated theoretically and experimentally the propagation of light along the optic axis of the cholesteric liquid crystals. The general agreement between theory and experiment confirm that the model that has been assumed is essentially correct. In this chapter we shall consider the propagation of light at right angles to the optic axis.

When light is incident normal at the screw axis of an homogeneous specimen of cholesteric liquid crystal, some interesting effects are observed. If the incident beam is parallel diffraction patterns are formed on a distant screen somewhat similar to those produced by an optical grating. These patterns were

observed by Sackmann, Meiboom, Snyder, Meixner and Dietz (1968). If the specimen is observed by through a microscope, alternate dark and (relatively) bright bands are seen. When the microscope is moved **eighter** closer or farther away from the speciman, there is a reversal in the pattern that is, the darker region becomes **brighter** and the brighter region dark. These were first observed by Robinson and Ward (private communication). We shall now discuss the theory of light propagation to the screw axis and explain these effects in some detail.

Subsequent work of R. Nityananda (M.C.L.C in print) has been shown that Chandrasekhar's model is equalant to the continuum MODEL When the thickness of the layers are very small.

In Chapter VI we discuss elobarately the applications of the cholesteric liquid crystal. We have discussed **fourteen** applications.

In the appendix we have discussed elabarately the Poincare Sphere and Jones Calculus.

CHAPTER I

OPTICAL PROPERTIES OF CHOLESTERIC LIQUID CRYSTALS PREVIOUS STUDIES

INTRODUCTION

The liquid crystalline or mesomorphic phase represents an intermediate state of matter between the crystalline solid and the isotropic liquid. It possesses some optical properties characteristic of crystals and simultaneously exhibits flow properties characteristic of liquids. Liquid crystals are broadly classified into three different types.

1. The smectic mesophase

The smectic mesophase is a turbid viscous state with certain properties reminiscent of those found in soaps. This phase consists of a layered structure wherein the molecules have their long axes parallel to one another (Fig. 1(a)). In smectic A phase the long axes are normal to the layers and there is no regularity of structure within each layer. If a regularity of structure exist within each layer, the phase is referred to as smectic B (Fig. 1(b)). In smectic C (Fig. 1(c)), the long axes of the layers are inclined to the layers, the angle of tilt being temperature dependent.

2. The nematic mesophase

The nematic mesophase is a turbid mobile state. This phase has average parallel orientation of molecules without any layered arrangement (Fig. 2(a)). A homogeneous specimen of the liquid crystal is uniaxial and optically positive.

3. The cholesteric mesophase

The cholesteric mesophase is a turbid and mobile mesophase. It is readily distinguished from the two other phases, because of its unique optical properties quite different from those of the other two. The molecular arrangement is of the nematic type with helical twist superimposed on it perpendicular to the long axis of the molecules. The structure may be described as a pile of thin birefringent layers, the principal axes of the successive layers turned through a small angle (Fig.2(b)).

In the first two or three decades following the discovery of the cholesteric liquid crystal several detailed studies on its optical properties were made by a number of workers, notably by Lenann (1889, 1890, 1900), Gaubert (1911, 1912, 1917), Giesel (1910), Vorlander and Huth (1913), Stumpf (1910, 1912, 1918), Grandjean (1912, 1917), Royers (1922, 1925) and Friedel (1922).

The cholesteric mesophase assumes various textures between plane glass surfaces, as observed in a microscope. Whenever a cholesteric compound is heated from the crystalline to the mesomorphic state, or cooled from the isotropic to the mesomorphic state, without any disturbance of the coverslip, one of the normal textures adopted is the so-called focal conic textures. In this texture the preparation contains an arrangement of fine dark lines which are observed both between crossed

nolaroids and in ordinary light. Neither the shape nor the arrangements of these lines is random; they are in fact ellipses hyperboles or portions of ellipses and hyperbole in association, such that the two lines are related as focal conics (Fig.3). The molecular arrangement will form a geometry such that the conics formed are perpendicular planes and each conic passed through the focus of the other (Friedel, 1922; Bragg, 1934). The optical sign of this texture is negative.

The second type of texture is called 'homotropic' wherein the mesophase appears in the form of a cloud which is not birefringent so that the field of view in the microscope appears dark between crossed polaroids. This texture is supposed to arise through rapid coalescing of a large number of small particles (Gray, 1962).

The third important texture obtained by a slight cover slip displacement over the slide containing either the 'homotropic' or the 'focal conic texture' is called 'Grandjean plane texture' (structure a plans). Here the molecules arrange themselves in parallel planes with a helical twist superimposed. It is this texture which exhibits the unique optical properties already referred to, which will be described in detail below.

The optical properties of the plane texture of cholesteric substances have been described in detail by Friedel (1922)

in his monograph and may be summarised as follows :-

- 1) The plane texture is uniaxial and optically negative.
- 2) When illuminated with white light, it scatters light to give vivid colours which, for a given substance, vary with angle of incidence and temperatures.
- 3) At normal incidence, it is strongly optically active with rotatory powers many times that of common optically active materials. It is also highly circularly dichroic for a certain wave length region; i.e. one circularly polarized component of incident beam is transmitted without attenuation whereas the other is reflected. Depending on the material, either the left or the right circular polarized component may be affected. The sense of circular polarization of the reflected light remains the same as the incident light. This is contrary to what is usually observed in the reflection from normal dielectrics.

These properties seem to depend on a delicately balanced molecular arrangement. Any disturbance that, mechanical, electrical etc., which interferes with the weak forces between molecular results in a dramatic change: reflection, transmission, birefringence, optical rotation and colour all undergo marked transformation. This property particularly the temperature sensitivity, has found extensive application in

recent years for non-destructive testing, mapping of skin temperatures, etc.

In this chapter, we shall review in detail the important experimental and theoretical investigations carried out on the propagation of light along the optic axis of cholesteric liquid crystals and discuss the points where the theories are unable to account for the observed facts satisfactorily.

EXPERIMENTAL STUDIES :

Stumpf (1910, 1912) was the first to make a systematic study of the optical rotatory power and reflection of a cholesteric liquid crystal. He measured rotatory power and extinction indices for circular polarised light as a function of wavelength for amyl-cyano-benzylidene-aminocinnamate, which though not a derivative of cholesterol shows all the characteristic properties of cholesteric substances. He observed that on either side of the wavelength of maximum selective reflection, the rotatory power of the medium varies in a manner somewhat similar to that observed on either side of an optically absorption band. Thus, the rotatory power is opposite sides of the wavelength of maximum selective reflection and increases as this wavelength is approached. No observations were recorded near the centre of the region of selective reflection. Stumpf's results are illustrated in figure (4).

Subsequently there were some studies on the optical properties of pure cholesteric liquid crystals and of mixtures of nematic and cholesteric compounds, but it was only in 1938 that detailed quantitative measurements of the rotatory dispersion right through the reflection band, the ellipticity of the transmitted light, and the circular dichroism, and of the effect of the temperature on these properties were carried out by Mathieu (1938). He used films of cinnamate, propionate, benzoate and pelargonate of cholesterol. The main conclusions drawn by Mathieu from his experiments are: 1) The reflected light at normal incidence is circularly polarized. The spectral width of the reflection is about 200 Å; the reflected intensity has a maximum at the centre of the band and diminishes rapidly on either side.

2) The transmitted light in this spectral region is elliptically polarized; the light becomes circularly polarized if the thickness of the cholesteric layer is sufficiently large.

3) The rotatory power never attains an infinite value, but decreased gradually as the wavelength approaches became very close to and then reverses sign on the other side.

The results are reproduced in the Figs. 5 and 6 for cholesteryl cinnamate.

The conclusions drawn by Mathieu were further confirmed by the studies of Levy (1948) on cholesteryl oleate and of Ferguson (1966) on mixture of benzoate, acetate and apmitate of cholesterol.

THEORETICAL STUDIES :

Mauguin's work :

A significant step in the understanding of the optical behaviour of the cholesteric substances was made by Mauguin (1911, 1913) in his attempt to explain an interesting feature of an experiment with a nematic liquid crystal, p-azoxyphenetole carried out by him. The same observations were made at about the same time independently by Lehmann (1912, 1916). He found that a nematic liquid crystal placed between the two glass plates sets a spiral twisting when the plates are rotated in opposite directions. This type of structure shows optical rotation of plane polarised light. Mauguin tried to explain this form of rotation by assuming an helically twisted birefringent medium using the Poincare sphere and rolling cone.

It is well known that light passing through an ordinary birefringent medium is transmitted as two plane polarized waves which have their electric vectors at right angles, to each other. These waves are called the 'proper waves' of the material [for a particular direction of propagation - Levelut And Billard (1970) had discussed the orthogonality of eigen waves (elastic and electromagnetic for transparent media, (homogeneous and twisted.)] Any wave whatever

incident on the medium must be resolved into two such proper waves and these waves are then propagated through the medium, independently of each other. The directions of the electric vectors of these two proper waves remain constant during their transmission of the medium for that particular direction. Mauguin assumes the principal directions to rotate continuously about the direction of propagation of the light as the medium is traversed. The Poincare sphere method used by Mauguin is a method of representing the azimuth and ellipticity of polarized light on the surface of a sphere.

In figure 7(a) the azimuth of the electric vector or plane waves propagated along the Z-direction is denoted by α and its ellipticity by β . This can be represented on the Poincare sphere as follows a-

Choosing any point A on the equator of the sphere marking of 2α from it round the equator and 3β round the meridian from the point thus reached, as in figure 7(b) a point P is arrived at, which represents Fig. 7(a). According to this notation, the equator represents all plane polarized light $\beta = 0$ and the poles circularly polarized light of opposite senses of rotation $\beta = \pi/2$, points at opposite end of a diameter in general represent ('crossed ellipses') i.e. ellipses of the same form with opposite senses of rotation and with their major axes at right angles. A pair of points at either ends of an equatorial diameter

represent a pair of plane polarized waves with their electric axes mutually perpendicular as (proper waves) of a birefringent medium.

Mauguin (1911, 1913) shows that the effect of light on a birefringent plate of thickness 'dt' and birefringence (Δn) is simply to rotate the point representing the incident light around an equatorial diameter taking it to some other point on the sphere, the ends of which represent the plane polarized proper waves of the plate, and the rotation to be given is

$$d\phi = \frac{2\pi (\Delta n) dt}{\lambda}$$

where λ = wavelength of light in vacuum.

The principal directions of the successive plates of the helically twisted birefringent layers in the stock will be represented by the points $A_1 A_2 \dots A_n$ and the diametrically opposite points $B_1 \dots B_n$ as in figure 8. Par plates of the same thickness

$$4 A_2 = A_2 A_3 \dots = A_n - 1 A_n = 2\alpha$$

The effect of this stack of birefringent layers on the incident light involves rotating the point P representing the incident light by an angle $d\phi$ around $A_1 A_1$ and by $d\phi$ around $A_2 B_2$ etc. successively. This can be done easily by considering a regular spherical polygon each side of which is of length $2d\alpha$ and whose interior angles are the supplement of $d\phi$ as follows : (A closed regular polygon

exists only if $m d\phi = 2\pi$ where m is a positive integer).

The spherical polygon is placed on the surface of the sphere so that the m th vertex lies on A_1 and one side lies on the equator to the right of A_1 . The point P representing the incident light is rigidly connected to the polygon and the polygon is allowed to roll round on the equator so that its vertices fall on $A_1 A_2 \dots A_n$. The polygon rotates about each point in turn and the locus of P is the required one (Fig.9).

$$\text{Now } d\phi = \frac{2\pi (\Delta n) dt}{\lambda} \quad \text{AND} \quad d\alpha = \frac{2\pi dt}{P}$$

Where P = pitch of helix.

Therefore $d\alpha/d\phi = \lambda/p (\Delta n)$ if $d\alpha$ and $d\phi \rightarrow 0$, keeping λ , p and Δn constant the polygon becomes a circle of angular radius 'w' where

$$\tan w = \frac{2}{p(\Delta n)}$$

(i.e. the circle subtends a cone of semiangle w at the centre of the sphere). Any point p on the equator and rigidly fixed to the cone will describe a path as in figure (1) as the cone rolls. The rate of rotation round the equator will be governed by the ratio of the circumference of the cap of the cone to the circumference of the equator. Let us the sphere have unit radius. Hence, its circumference will be 2π .

The circumference of the cap is $2\pi \sin w$ since the radius of the cap is $\sin w$.

$$2\pi \sin w = 2\pi \tan w (1 + \tan^2 A)^{-1/2} = 2\pi \left[1 + \frac{p^2 (\Delta n)^2}{4 \lambda^2} \right]^{1/2}$$

A distance $P/2$ along the Z axis corresponds to a structural twist of π radians. For this amount of twist the cone will make one complete revolution around the equator * in the opposite sense through an angle

$$\theta = 2\pi \left[1 - \left\{ 1 + \frac{(\Delta n)^2 p^2}{4 \lambda^2} \right\}^{-1/2} \right] \quad \dots(1)$$

θ is the optical rotation and has a negative sign because it is in the opposite sense to the structural twist.

Now if $(\Delta n)^2 p^2$ is small compared to $4 \lambda^2$ expanding the root and retaining only first powers of $(\Delta n)^2 p^2 / 4 \lambda^2$ gives $\theta = -\pi (\Delta n)^2 p^2 / 4 \lambda^2 \quad \dots(2)$

The optical rotation per unit thickness will have

$$\theta = -\pi (\Delta n)^2 p^2 / 4 \lambda^2 \text{ radians} \quad \dots(3)$$

Neglecting the dispersion of birefringence, we see from (3) that the rotatory power versus the reciprocal of the square of the wavelength gives a straight line.

* and the representative point P will move around the equator.

Experimental verification of Mauguin's formula :

The first attempt to verify quantitatively Mauguin's theory was by Robinson and Ward (1957). They discovered that solutions of some polypeptides in certain organic solvents for example poly- γ -benzyl-L-glutamate (PBLG) in dioxan, methylene chloride, chloroform and m-cresol at certain concentrations spontaneously become birefringent. Under suitable conditions, the solutions exhibited equi-spaced alternate dark and light lines when observed through microscope. The lines may be interpreted as a view of the structure at right angles to the helical axis. Robinson confirmed this interpretation by observations between crossed polaroids and also by use of quartz wedge. The retardation plotted against distance in a direction at right angles to the lines had an oscillating value thereby confirming that expected from the structure. The large spacings of the polypeptides could be conveniently varied by altering the concentrations of the solvent. When view along the helical axis no lines were seen, but a very high optical rotatory power was present. They measure the optical rotatory power various wavelengths for fourteen different solutions covering a range of both concentration and solvents and thereby for P values ranging from 10-200. μ . In all cases the proportionality to λ^2 was found to hold over the visible spectrum. This has been reproduced for PBLG in chloroform in figure (11).

Next they substituted the values of rotatory power and pitch in the Mauguin's equation (3) and calculated the layer birefringence per volume fraction of the polypeptide in solution. It turned out as 0.026 and was remarkably constant despite the varying values of ρ and p . Then they prepared a racemic mixture containing equal quantities of the D and L enantiomers (PBDG and PBLG) which too under certain condition adopted the spontaneously birefringent phase. Only, in this case it was not the twisted cholesteric structure, but the untwisted nematic structure. They measured the birefringence directly and obtained the birefringence per volume fraction as 0.029. This was reproducible in many cases. Thus the experiments of Robinson and Ward show quite conclusively that the model is an accurate representation of the structure of the cholesteric liquid crystal.

Chatelain and Cano(1961, 1969) have made extensively measurements of rotatory power on mixtures of cholesteric and nematic substances. Whenever a small quantity of the cholesteric substance or even a non-mesomorphic optically active substance is added to a nematic compound it twists the structure to form a cholesteric type of liquid crystal. They have measured rotatory dispersion of a mixture of cholesteric benzocyclobutene and p-azoxyphenetole for various concentrations and temperatures. They have also calculated rotatory power according to Mauguin's formula (3) for various ρ

wavelengths using the experimental values of the pitch and assuming the layer birefringence of the mixture to be that of p-azoxyphenetole. This assumption is valid to a good approximation, since the birefringence of the p-azoxyphenetole is very large compared to that of cholesteric benzoate and the small quantity of the latter compound affects the birefringence of the mixture only very slightly. Tables of the measured values and calculated values of rotatory dispersion are given in Tables I and II for a mixture 5/100 of cholesteric benzoate and p-azoxyphenetole.

Table II a - Measured values of rotatory dispersion for a mixture 5/100 of cholesteric benzoate and PAP (chatelain and ceno, 1984)

(A)	8438	5791	5461	5086	21916	4678
t°C						
155	18000°	24100°	29200°	38700°	45500°	59000°
150	22000°	28500°	34700°	45200°	53100°	68600°
145	24600°	32200°	38100°	50000°	59000°	76200°
140	26300°	35000°	42500°	56000°	65600°	84000°
135	28300°	37600°	45500°	60400°	70800°	90000°
130	29900°	40400°	48900°	65200°	76800°	95200°
125	31900°	42600°	52500°	69300°	81000°	102000°

Table 2 : - Calculated values of rotatory dispersion for a mixture 5/100 of cholesteric benzoate and PAP (Chatelained cano, 1964).

(A)	6438	5791	5461	5086	4916	4673
$t^{\circ}\text{C}$						
155	16500°	23600°	29500°	38700°	47600°	63200°
150	19400°	27800°	34900°	47000°	56000°	74900°
145	21900	31700	39700	53700	63600	85100
140	23800	34100	42600	57900	69000	91900
135	25500	36200	45100	61900	73800	98200
130	26800	37900	47400	65400	78100	103000
125	28800	39500	49200	68000	81800	108000

The experiments of Robinson and Ward and of Chatelain and Cano show quite conclusively *thht* the Mauguin's model is a satisfactory description of the cholesteric structure. The Mauguin's theory not, however, take into account the effect of reflections the interface between the layers which are bound to occur when the wavelength of the radiation in the medium becomes comparable to the pitch of the helix. It is unable to explain the anomalous rotatory dispersion round a region

of reflection as observed by various workers.

In principle the general problem can be solved by an electromagnetic treatment, of which several have been given in the literature. One such is by Oseen (1933) who tries to explain by conjecturing a helicoidal structure for the stratified arrangement of the Grandjean plane texture. This model was previously proposed by Friedel (1922) in drawing the similarities between cholesteric and nematic substances. The theory is presented in very general terms so that no verification has been made. Another theory was developed by de Vries (1951), which succeeded in giving a broad explanation of the relation between reflection and rotatory power.

de Vries' theory :

de Vries derives an equation relating the optical rotatory power to the pitch of the helix, the wave length of light and the birefringence taking also into account the selective reflections at the interfaces between the layers, which plays an important role when the pitch of the helicoidal structure of the order of the wavelength of light on the medium.

de Vries first assumes the liquid crystal to consist of a large number of thin anisotropic layers, the principal axes of the successive layers turned through an angle β .

It is well known that reflection at the boundary of two layers which make optical can occur only when there is a difference 3% the refractive indices at the interface. If right circularly polarized light be incident along the screw axis of a right handed structure, then resolving the wave into linearly polarized components, both linear components will experience a change of refractive index when passing from one layer to the next. The effect of this is that one linear component will be reflected with phase reversal and the other without phase reversal. Therefore viewed along the direction of propagation of the reflected light, the sense of circular polarization remains the same as that of the incident light. This is contrary to what is observed in reflection from normal dielectrics.

Now circularly polarized light may be regarded as one in which the tip of the electric vector describes a spiral path as it is propagated. It is evident that if the pitch of this helix is exactly the same as that of the helical structure, reflection from successive layers will take place under identical circumstances, so that the reflected waves from the successive layers will constructively interfere to give rise to a maximum. In other words the condition for reflection is $\lambda = pn$, where $n =$ mean refractive index of the medium and $p =$ pitch

of helix. On the other hand if left circular light be incident on the right handed structure, such an interference maximum will not occur. Thus the circular component will be strongly reflected, while the other will be transmitted without change.

An expression for the rotatory power is obtained as follows :

According to Fig.(12) X and Y are laid along the electrical axes of the first layer, and ξ , η axes are along the corresponding directions of the second layer which is turned through an angle θ . Let the normally incident linearly polarized wave be $e_x \exp 2\pi i(t/T - nx/\lambda)$ with its electric vector making an angle θ with respect to X axis. Transferring this equation into ξ and η axes, the relations between them are

$$E_X = E_\xi \cos 2\pi x/p - E_\eta \sin 2\pi x/p$$

$$E_Y = E_\xi \sin 2\pi x/p + E_\eta \cos 2\pi x/p$$

where p = pitch of helix.

Inserting the solution	$\begin{array}{l} E_\xi = A \\ E_\eta = iB \\ E_z = 0 \end{array} \left \begin{array}{l} \\ \\ \end{array} \right. \exp 2\pi i(t/T - nx/\lambda)$
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de Vries obtains for A and B the linear simultaneous equations

$$\begin{array}{l} A \left(\frac{\epsilon_1}{\lambda^2} - \frac{m^2}{\lambda^2} - \frac{1}{p^2} \right) - \frac{2Bm}{p} = 0 \\ B \left(\frac{\epsilon_2}{\lambda^2} - \frac{m^2}{\lambda^2} - \frac{1}{p^2} \right) - \frac{2Am}{p} = 0 \end{array} \quad (4)$$

ϵ_1 and ϵ_2 are principal dielectric constants along and . These are consistent if the determinant of the coefficients of A and B vanishes giving a quadratic equation for m^2 .

Introducing the quantities

$$\lambda' = \frac{\lambda}{p \sqrt{\epsilon}}; \quad m' = \frac{m}{\sqrt{\epsilon}}; \quad \alpha = \frac{\epsilon_2 - \epsilon_1}{2\epsilon}$$

where $\epsilon = \frac{\epsilon_1 + \epsilon_2}{2}$

the quadratic terms out as

$$m'^4 - 2m'^2 (1 + \lambda'^2) + (1 - \lambda'^2)^2 - \alpha^2 = 0$$

and equation 4 reduces to

$$B/A = \frac{1 - \alpha - m'^2 - \lambda'^2}{2m'}$$

Expanding in series the two roots of the equation

$$m'_1 = 1 - \lambda' - \frac{\alpha^2}{8\lambda'(1-\lambda')} + \dots$$

$$\frac{m'}{2} = 1 + \lambda' + \frac{\alpha^2}{8 \lambda' (1 + \lambda')} + \dots$$

do Vries shows that the proper waves outside the region of reflection are circularly polarized with opposite signs of rotation and having different velocities. This brings about a rotation of the plane of polarization of a linearly polarized beam which can be considered as resulting from these normal waves. The direction of the resulting vector is the bisector of the angle between simultaneous positions of the two rotating vectors. Calling the angles of these three vectors with the X rotating axis θ , ϕ , and ψ respectively, the rotating power is

$$d\psi/dz = 1/2 \left(\frac{d\psi_1}{dz} + \frac{d\psi_2}{dz} \right)_t$$

Now $(d\psi/dz)_t$ consists of two parts: its rotation in the $\xi - \eta$ system and rotation of the system itself.

One finds

$$d\psi_2/dz = \frac{2\pi}{p} (1 - m'_2/\lambda') \quad \text{and} \quad d\psi_1/dz = \frac{2\pi}{p} (1 + m'_1/\lambda')$$

Inserting

$$\frac{m'_1}{1}, \quad d\psi_1/dz = 2\pi/p \lambda' \left(1 - \frac{\alpha^2}{8 \lambda' (1 - \lambda')} + \dots \right)$$

$$d\psi_2/dz = 2\pi/p \lambda' \left(1 + \frac{\alpha^2}{8 \lambda' (1 + \lambda')} + \dots \right)$$

and simplifying

$$d\psi/dx = \frac{-2\pi}{p} \frac{\alpha^2}{8\lambda^2(1-\lambda^4)}$$

AND REFLECTION WILL BE SIGNIFICANT INSIDE THE NARROW LIMITS

$$\lambda \pm \frac{\lambda\alpha}{2}$$

where $\alpha = (n_2 - n_1)/n$, where $n_1 - n_2$ is the lower birefringence and $n = (n_1 + n_2)/2$

This expression for rotation is identical with the one derived by Hauguin's rolling cone method except for the factor $(1 - \lambda^2)$ in the denominator which results from the reflection and reduces to unity for large values of the pitch i.e. when $\lambda \ll p^{1/2}$.

Fig (13) shows the reflection and rotatory power as a function of the reduced wavelength for some typical values of the constants reduced wavelength λ for some typical values of the constants.

$$\alpha = 0.05; \quad p = 0.4$$

We see from the curves that the rotatory power shoots up to a finite maximum value, reverses sign on the sides of the reflection band and decreases gradually.

de Vries has also discussed the theory of the reflection at the boundaries of the liquid crystal, and as well as the reflection from thin layers. He has shown

theoretically that the reflected intensity for a thin specimen is a function of the thickness d and extends beyond the region of total reflection from an infinitely thick liquid crystal.

The theory in its present form does not make any prediction about the rotatory power inside the region of reflection whereas, we have seen earlier that experiments show that the rotatory dispersion inside the region of reflection ties up smoothly with the rotatory dispersion curve outside the region of reflection.

In the next chapter we discuss a new approach put forward by Chandrasekhar and Srinivasa Rao (1968) for explaining the optical properties of cholesteric liquid crystal for light propagation on along the optic axis. ~~The theory is presented here with some modifications and~~ improvements. We shall in the next chapter extend the theory of films of finite thickness, as encountered in actual practice.

In next Chapter we shall give some detailed experimental data on rotatory power, reflection. As we shall see, the theoretical predictions are in agreement with the observed phenomena.

In the next chapter we shall consider the propagation of light at right angles to the optic axis.

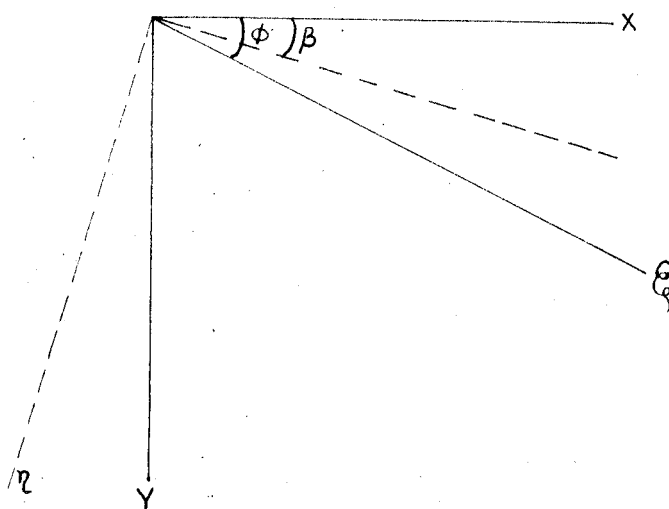


FIG. 12 Representation of the electric axes of first two layers

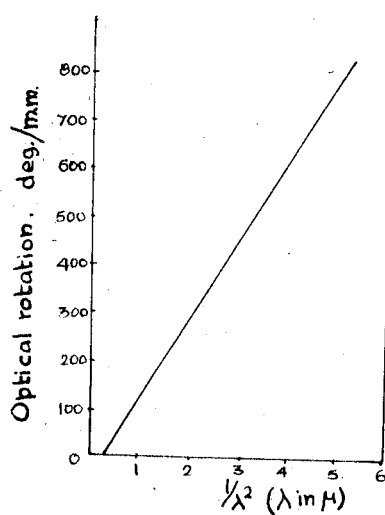


FIG. 11 Dependence of optical rotation on wave length PBLG in chloroform. $18.9/100g$, $S = 2 \mu$ (Robinson and Ward - 1957)

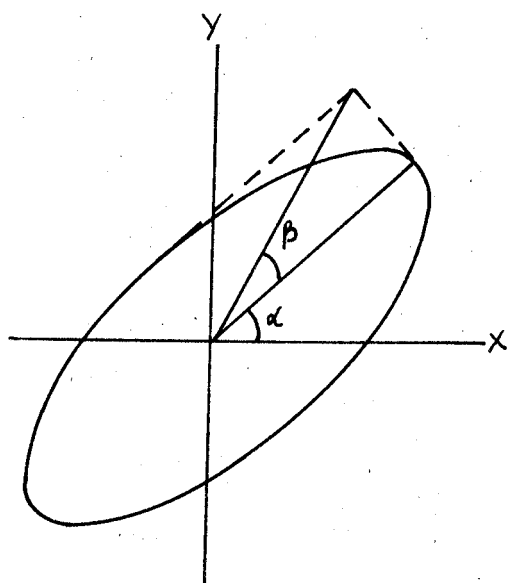


FIG. 7a. Elliptically polarised light.

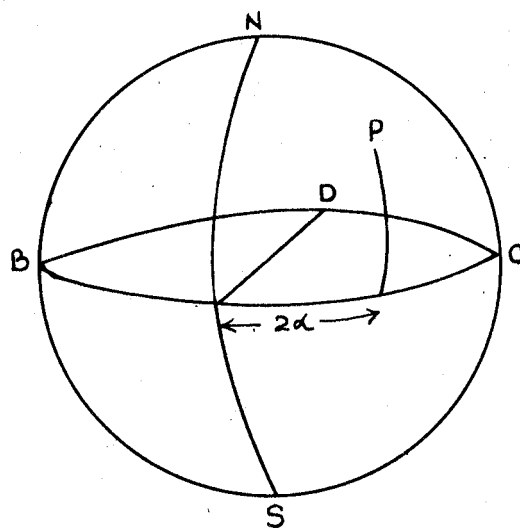


FIG. 7b. Representation of above on a poin care' sphere

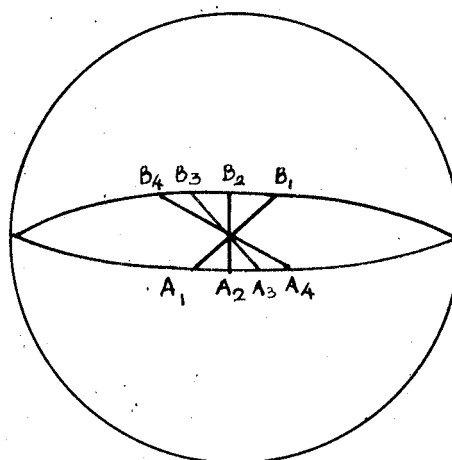


FIG. 8 Principle directions of Successive plates of helically twisted birefringent layers

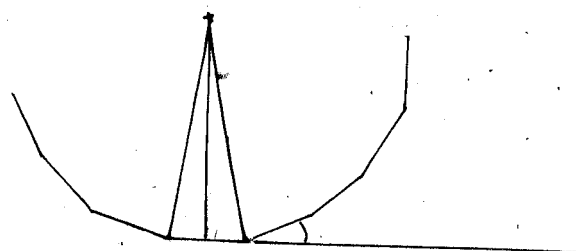


FIG. 9 A regular spherecally Polygon

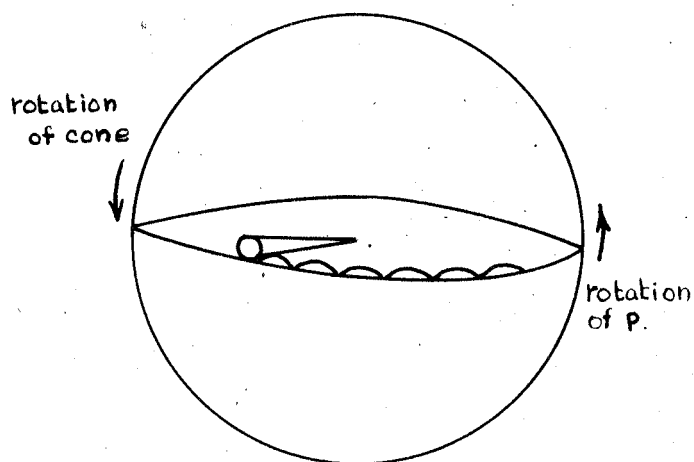


FIG. 10 The path described by a path P on the equator as the cone rolls.

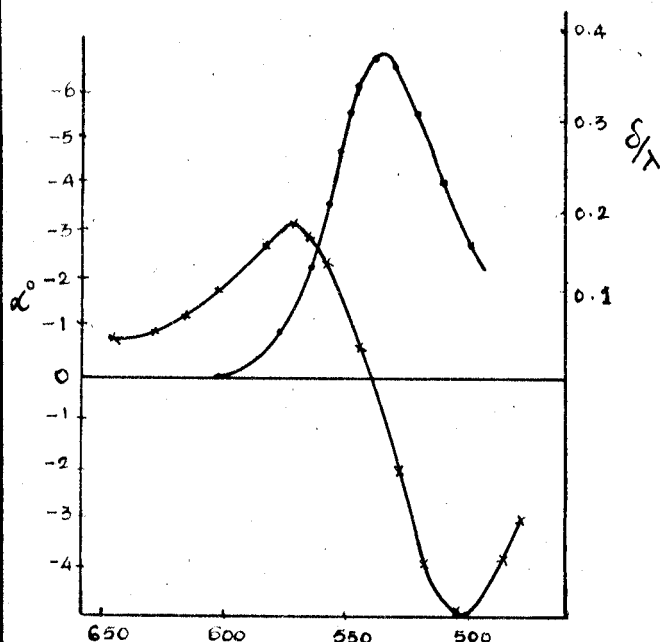


FIG.5 Rotatory dispersion and ellipticity in cholesteryl cinnamate (Mathieu 1938)
X. Rotation; Ellipticity -
Cinnamate de cholestérine. $t \sim 50'$

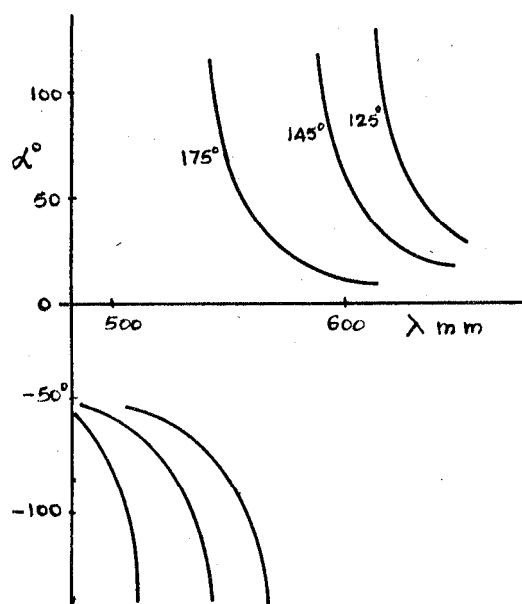


FIG.6 Optical rotation as a function of λ in cholesteryl cinnamate (Mathieu 1938)

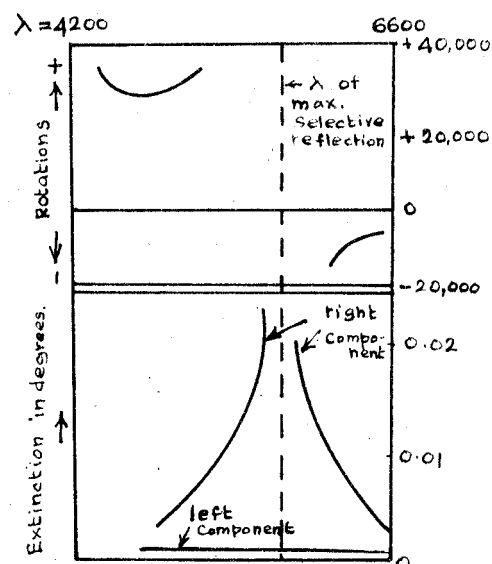


FIG.4 Optical activity in liquid crystals Amyl - cyanobenzylideneamino - cinnamate (Stumpf 1910)
The right circular polarized component is selectively reflected, whilst, the left circular component is transmitted. The λ s increase from left to right.

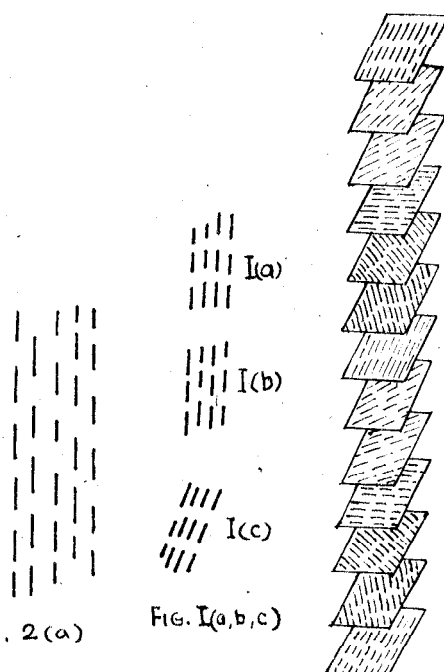


FIG. 2(a)

FIG. I(a,b,c)

FIG. I, 2b.

FIG I Structure of cholesteric phase

FIG I(a,b,c). Schematic representation of molecular arrangement in the three meso phase
Schematic A; Ib: Schematic B; Ic: Schematic C.

FIG. 2a. Nematic

FIG.2b Cholesteric

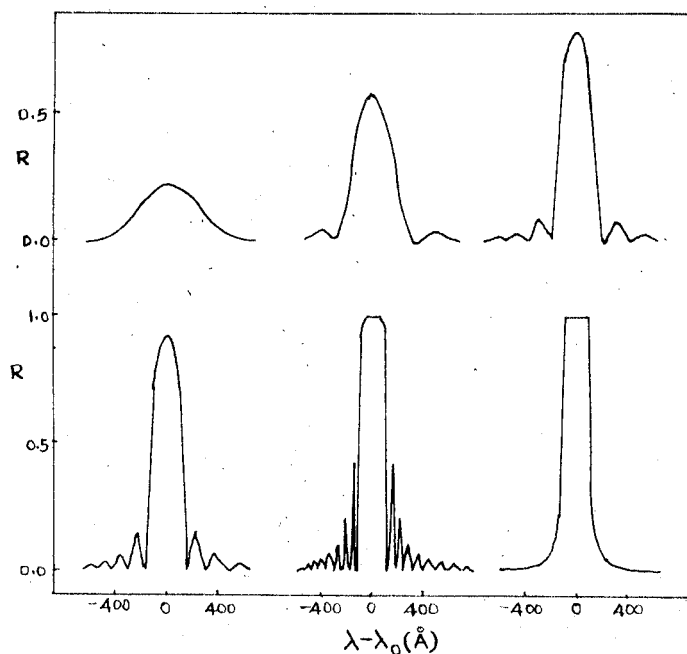


FIG.2 Theoretical reflexion curves for films of thickness mP ;
 $m = 5, 10, 15$ (top row), $20, 40$ and ∞ (2nd row).

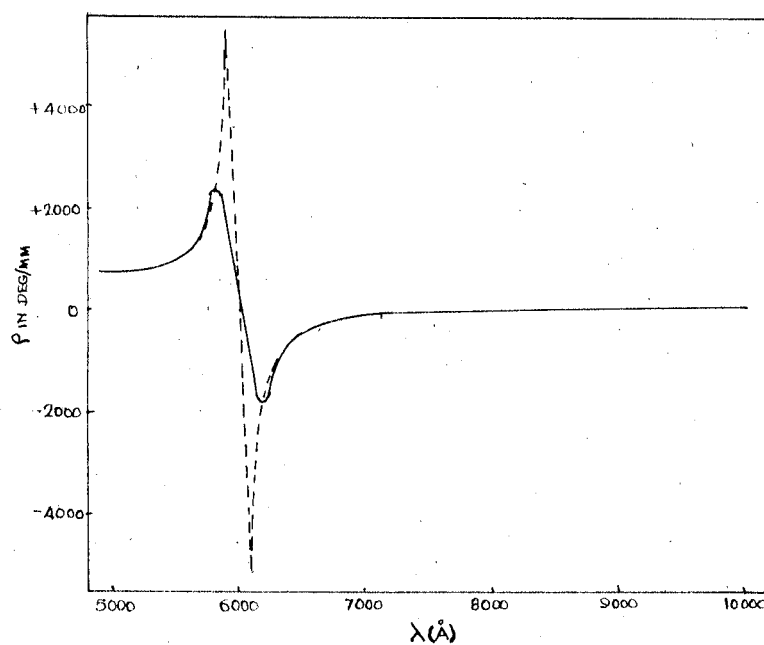


FIG.3 Theoretical rotatory dispersion curves; $m = 10$ (full curve),
 $m = \infty$ (broken curve).

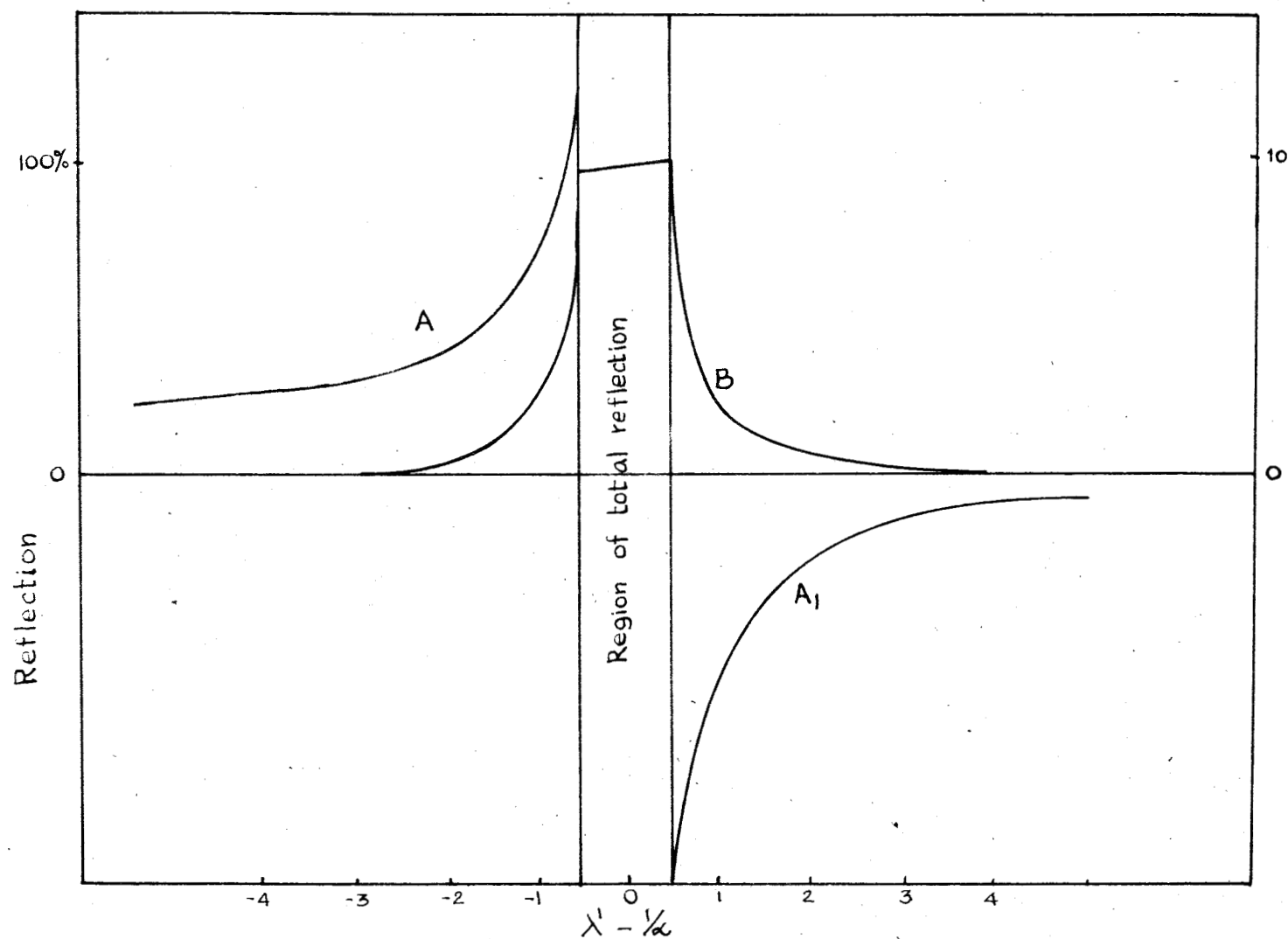


FIG. 13. Reflection and rotatory power as a function of reduced wave length λ' (de Vries-1951)