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Selective Reflection by Cholesteric Liquid Crystals†

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Abstract—Selective light reflection at the plane structure of a cholesteric liquid crystal is treated by the theory of de Vries[1] for normal incidence only. A more general approach is outlined for arbitrary angles of incidence. Conditions for the borders of the reflection bands are given. Also the cases of ordinary total reflection are included. Using the derived formula, the two principal dielectric constants are determined from the angles of total reflection measured with a refractometer. Their values are used to calculate the reflection spectrum of a layer of finite thickness at normal incidence which is compared with an experimental spectrum. In agreement with the experimental curve the calculated spectrum shows a central band with a nearly rectangular shape flanked by a series of maxima and minima due to interferences, the positions of which depend strongly on the dielectric constants.

General Theory

The optical properties of cholesteric liquid crystals are described very well by the theory of de Vries[1] but only for light normally incident on a plane structure. Recently Taupin[2] and Berreman and Scheffer[3] treated the case of oblique incidence and calculated by numerical techniques reflected and transmitted intensities. The latter two authors were able to calculate for a given sample practically exact intensity curves as a function of wavelength. The angle of incidence was assumed to be 45° and two cases of different polarizations of the incident light were considered. In the first part of this paper we outline a different approach which allows a direct calculation of the

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borders of the reflection bands as a function of the angle of incidence.

We adopt Oseen's(4) model and assume the cholesteric liquid crystal to have a uniformely twisted nematic structure, the helical axis being perpendicular to the nematic symmetry axis. Accordingly we describe the dielectric properties by a tensor of rotational symmetry that changes the orientation with the twist.

For the treatment of light propagation in this system it is convenient to use two coordinate systems (Fig. 1): the laboratory system x, y, z with the z-axis along the helical axis and a local coordinate system ξ, η, ζ with the ζ-axis parallel to the z-axis. The other two axes are positioned in such a way that the dielectric tensor is diagonal in it with principal values ϵ_1, ϵ_2, ϵ_3 along the axes ξ, η, ζ. Thus, the electric field **E** and the dielectric displacement **D** are connected by the relations

$$\begin{aligned} D_\xi &= \epsilon_1 E_\xi \\ D_\eta &= \epsilon_2 E_\eta \\ D_\zeta &= \epsilon_3 E_\zeta . \end{aligned} \tag{1}$$

We introduce a unit vector **L** parallel to the nematic symmetry axis. Because of the uniform twist its components in the laboratory system are given by

$$\mathbf{L} = \{\cos\alpha,\ \sin\alpha,\ 0\} \tag{2}$$

where $\alpha = 2\pi\, z/p$ and p the pitch of the helix.

The problem of wave propagation consists in solving the Maxwell equations or the correspondent wave equation

$$\frac{1}{c^2}\frac{\partial^2 \mathbf{D}}{\partial t^2} = \Delta\mathbf{E} - \operatorname{grad}\operatorname{div}\mathbf{E}. \tag{3}$$

We assume that light passes from an isotropic medium of refractive index n into the liquid crystal (Fig. 2). Let the angle of incidence be ψ. The helix axis is taken to be normal to the interface.

Without loss of generality we confine ourselves to waves propagating in the (x, z)-plane and try to find solutions of the form

$$\mathbf{E} = \mathbf{F}(z)\exp\left[i\omega\left(t - \frac{m}{c}x\right)\right] \tag{4}$$

with $m = n\sin\psi$ and $F(z)$ an unknown vectorfunction. The components of $F(z)$ are given in the laboratory system by

$$\mathbf{F}_{lab}(z) = \{F_x, F_y, F_z\} \tag{5}$$

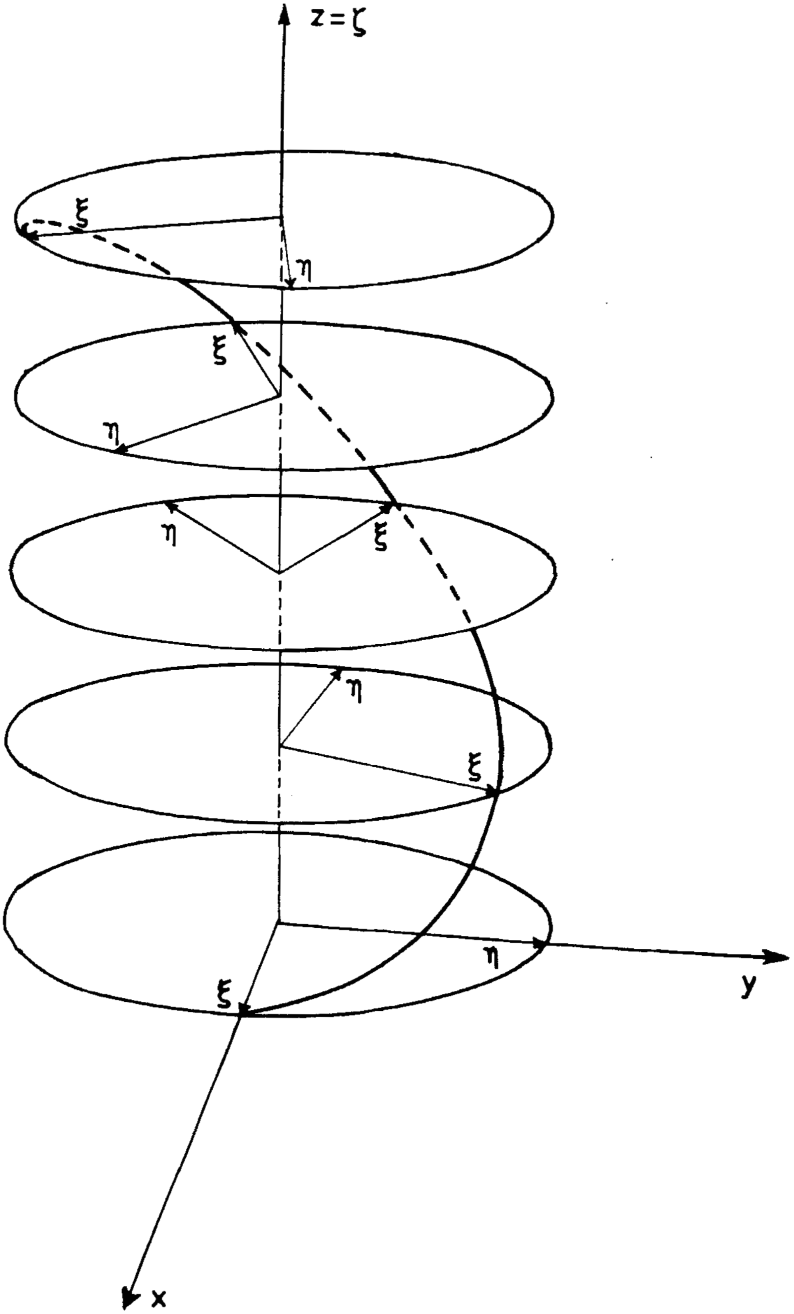

Figure 1. Definition of the laboratory and local coordinate systems.

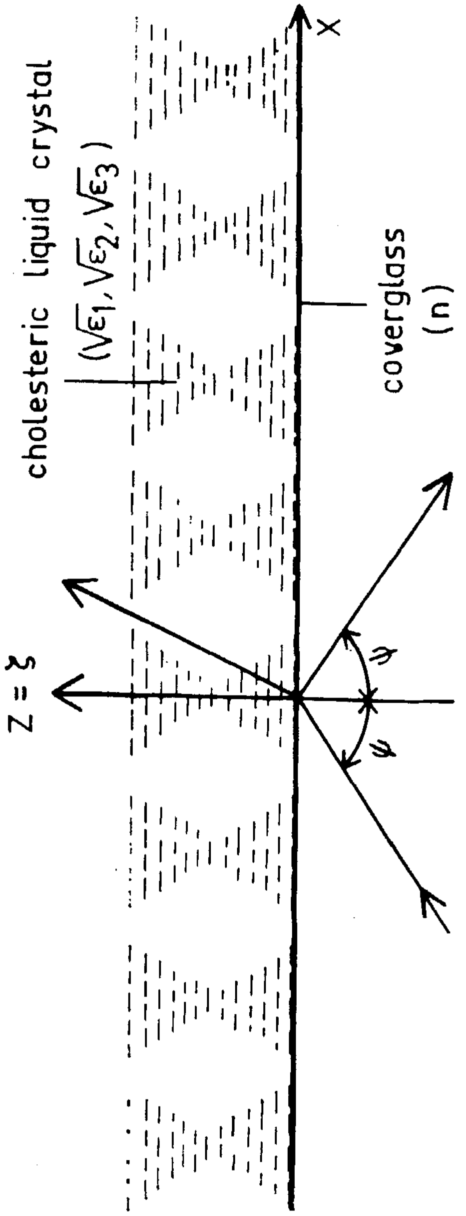

Figure 2. Light propagates in the (x, z)-plane from an isotropic medium of refractive index n into the liquid crystal.

and in the local system by

$$\mathbf{F}_{loc}(z) = \{\phi, \chi, F_z\}. \tag{6}$$

We chose the ξ axis parallel to **L** and have because of the nematic symmetry

$$\epsilon_2 = \epsilon_3. \tag{7}$$

We obtain for ϕ and χ a system of second order differential equations with periodic coefficients:

$$\begin{aligned} &\frac{d^2\phi}{d\alpha_2} - 2\frac{d\chi}{d\alpha} + (a_1 + a_2 \cos 2\alpha)\phi = 0 \\ &\frac{d^2\chi}{d\alpha^2} + 2\frac{d\phi}{d\alpha} + b_1\chi - a_2 \sin 2\alpha \cdot \phi = 0. \end{aligned} \tag{8}$$

For the third component we obtain

$$F_z - i\frac{\lambda}{p}\frac{m}{\epsilon_2 - m^2}\frac{dF_x}{d\alpha} = 0 \tag{8'}$$

where $\alpha = 2\pi z/p$, and

$$\begin{aligned} a_1 &= \left(\frac{p}{\lambda}\right)^2\left[\epsilon_1 - \frac{m^2}{2}\left(\frac{\epsilon_1}{\epsilon_2} + 1\right)\right] - 1 \\ a_2 &= \left(\frac{p}{\lambda}\right)^2\frac{m^2}{2}\left(1 - \frac{\epsilon_1}{\epsilon_2}\right) \\ b_1 &= \left(\frac{p}{\lambda}\right)^2[\epsilon_2 - m^2] - 1. \end{aligned}$$

We can also consider instead of the system (8) a single fourth order differential equation with periodic coefficients:

$$\frac{d^4\phi}{d\alpha_4} + (a + a_2 \cos 2\alpha)\frac{d^2\phi}{d\alpha^2} - 6a_2 \sin 2\alpha\frac{d\phi}{d\alpha} + (b + da_2 \cos 2\alpha)\phi = 0 \tag{9}$$

where $a = 4 + a_1 + b_1$

$b = a_1 b_1$

$d = b_1 - 8.$

According to Floquet's Theorem we expect solutions of the form

$$\phi(\alpha) = e^{i\mu\alpha}P_1(\alpha); \qquad \chi = e^{i\mu\alpha}P_2(\alpha). \tag{10}$$

P_i is a periodic function with period π. μ is the characteristic exponent. Since P_i is periodic and therefore bounded, the characteristic exponent decides if wave propagation is allowed or forbidden depending on a real or complex value. It can be shown that on the borders of the reflection bands μ is an integer:

$$\mu(\lambda, \psi) = \nu, \qquad \nu = 0, \pm 1, \pm 2, \ldots \tag{11}$$

The characteristic curves defined by Eq. (11) can be calculated relatively easily.

They describe the shift and splitting of the main reflection band with increasing angle of incidence and the occurence of higher order bands. This will be shown in detail in a later paper.

One characteristic curve is given by the simple equation

$$\lambda = p\sqrt{\epsilon_2 - m^2}, \qquad m = n \cdot \sin\psi. \tag{12}$$

It corresponds to the solution $\phi = 0, \chi = \text{const}, F_z = -i(P/\lambda)m\chi \cos\alpha$. An equation similar to (12) has been used by Fergason$^{(5)}$ for the center of the reflection band. He derived his equation treating the reflection like a simple Bragg reflection.

Determination of the Dielectric Constants

The characteristic curves give the conditions for the selective reflection as well as for the ordinary total reflection. For ordinary total reflection the following two relations were derived:

$$\begin{aligned} m_1^2 &= \epsilon_3 \\ m_2^2 &= \epsilon + \frac{\delta^2}{32(\lambda^2/p^2)}\left(1 - \frac{\delta}{\epsilon}\right) + o\left[\left(\frac{\delta}{\epsilon}\right)^4\right] \end{aligned} \tag{13}$$

$m_1 = n \sin\psi_1$ and $m_2 = n \sin\psi_2$, where ψ_1, ψ_2 are the angles of total reflection as measured with a refractometer, $\epsilon = \frac{1}{2}(\epsilon_1 + \epsilon_2), \delta = \frac{1}{2}(\epsilon_1 - \epsilon_2)$.

The first relation shows that m_1 is identical with one of the dielectric constants, ϵ_3. This relation remains valid for cases of lower symmetry where $\epsilon_3 = \epsilon_2$. It is assumed for the second Eq. (13) that $\lambda^2 > p^2\epsilon/8$. In most cases m_2 gives in a good approximation the mean value ϵ and does not correspond directly to a principle dielectric constant. This fact was not realized by earlier workers (see for instance Ref. (6)).

Figure 3 shows a plot of the two dielectric constants versus wavelength determined using Eq. (13) for a mixture of cholesteryl nonanoate, cholesteryl chloride and cholesteryl acetate in weight ratios of 20:15:6 at room temperature (24 °C).

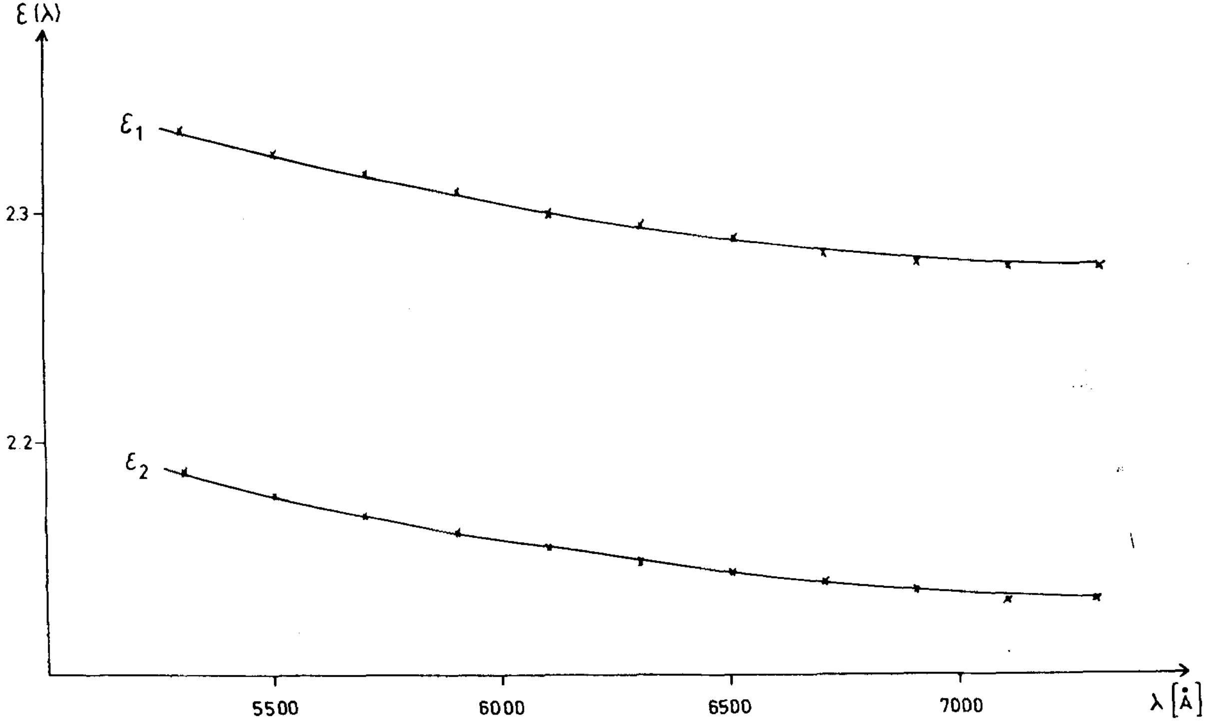

Figure 3. Local dielectric constants of a mixture of cholesteryl nonanoate, cholesteryl chloride and cholesteryl acetate in weight ratios of 20 : 15 : 6 at 24 °C.

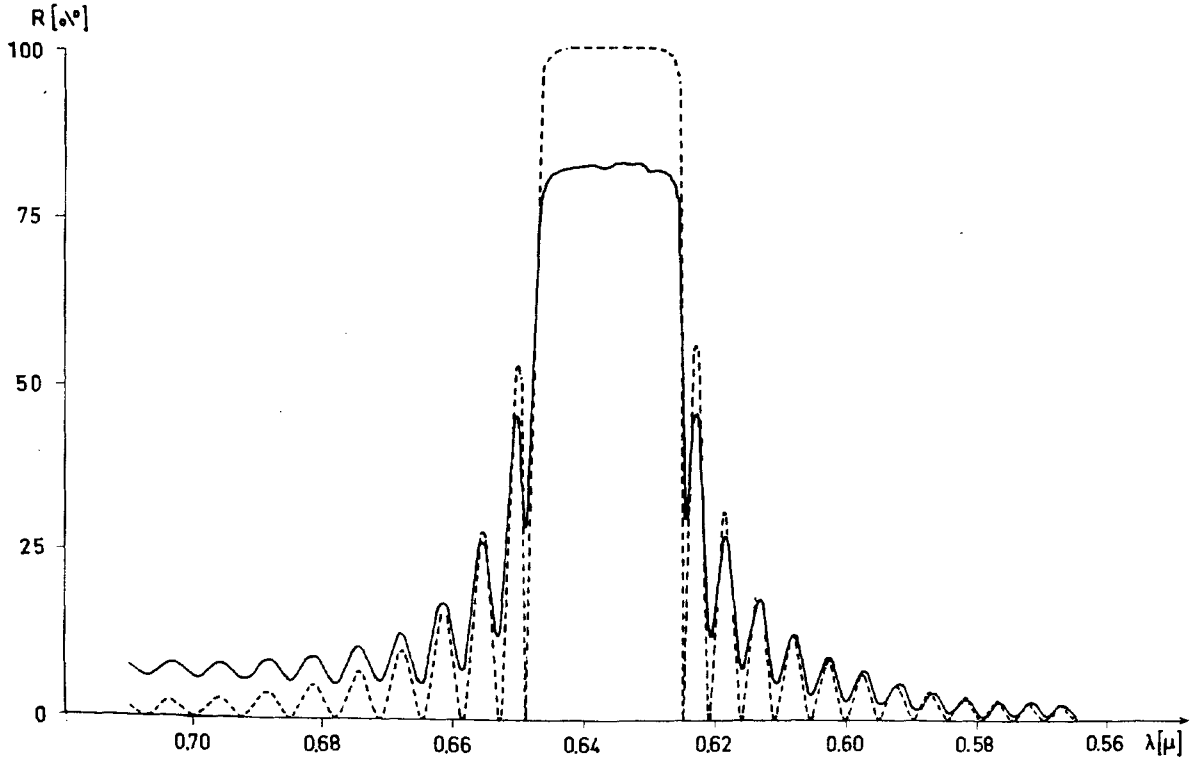

Figure 4. Reflection spectrum at normal incidence. Dashed curve: computed spectrum with pitch $p = 0.4273\mu$ and thickness of layer 21.0μ. Solid curve: experimental spectrum (intensity in arbitrary units).

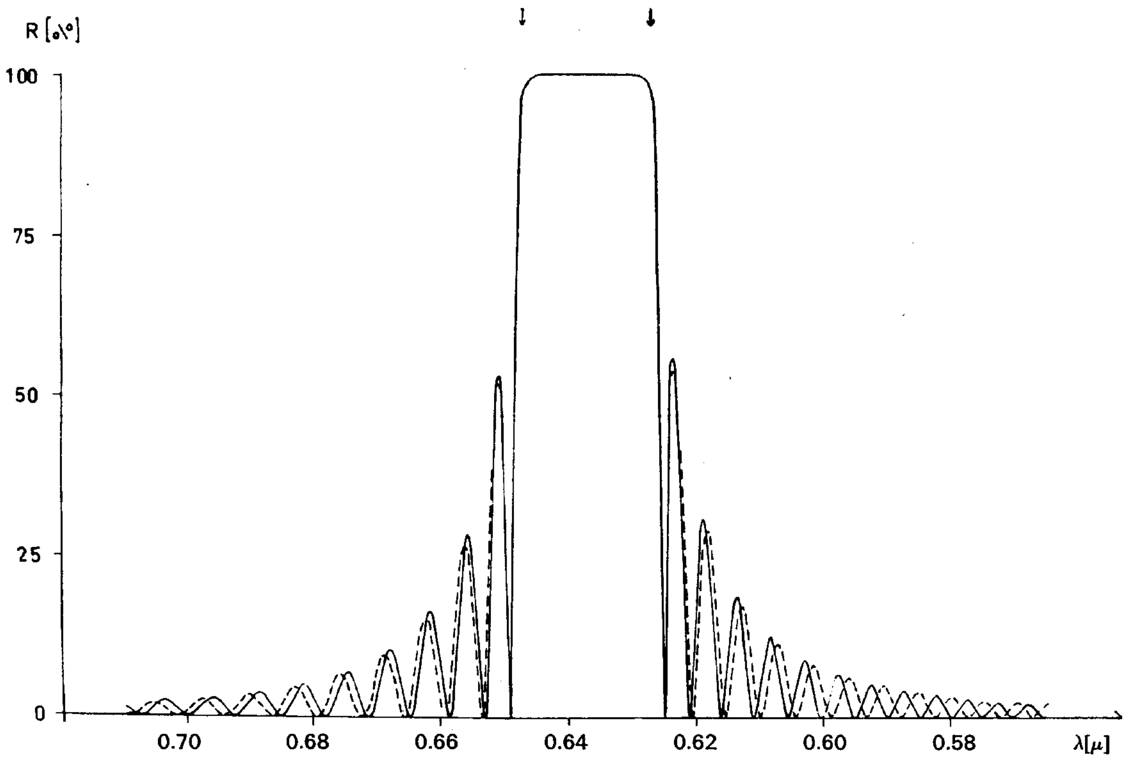

Figure 5. Computed reflection spectra at normal incidence. Solid curve: same spectrum as in Fig. 4. Dashed curve: neglecting the wavelength dependence of the dielectric constants results in a remarkable shift of the positions of the interference lines.

Reflection Spectrum at Normal Incidence

Figure 4 shows the reflection spectrum of the above mentioned mixture at room temperature for normally incident light. The solid curve gives the experimental result in arbitrary units. The dashed curve was calculated using the values of the dielectric constants shown in Fig. 3. The pitch was determined from the center of the reflection band using de Vries' formula $\lambda_0 = n_0 p$ with $n_0 = \frac{1}{2}(\sqrt{\epsilon_1} + \sqrt{\epsilon_2})$.

Both curves are in agreement concerning:

(i) the nearly rectangular shape of the central band,
(ii) the decreasing amplitude of the secondary minima and maxima with increasing distance from the region of reflection and
(iii) the position of the minima and maxima.

The positions of the minima and maxima depend strongly on the values of the dielectric constants used. Thus, the comparison of the measured and calculated spectrum represents a test of our method of determining the dielectric constants.

The strong dependence of the positions of the interference lines on the dielectric constants is demonstrated in Fig. 5. Here constant values of the dielectric constants were used neglecting their weak wavelength dependence. There results a remarkable discrepancy. Fitting the curves by varying other parameters, e.g. the thickness of the layer is not possible. There is also another point of interest. The dielectric constants that can be determined by total reflection in a refractometer are ϵ_3 and approximately ϵ. With these values and assuming $\epsilon_2 = \epsilon_3$ we calculated the reflection spectrum for normally incident light which depends on [illegible] and ϵ_2 only. We consider the good agreement of the theoretical and experimental curves as a strong confirmation of the assumption that the local dielectric properties are uniaxial.

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