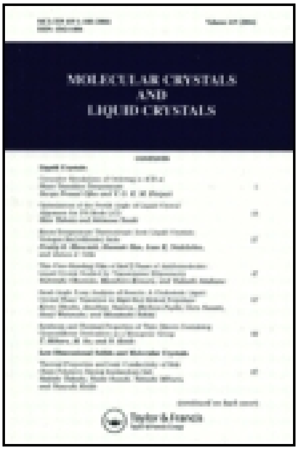

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Dispersive Reflection in Cholesterics

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Abstract—The general case of dispersive reflection in cholesteric liquid crystal films is treated. An expression is derived which relates angles of incidence and observation to film parameters. Experimental verification over a wide range of angular conditions is presented.

1. Introduction

Fergason[1] has shown that dispersive reflection from the Grandjean (plane) texture of the cholesteric mesophase can be treated in terms of Bragg like scattering from sites imbedded in a medium with refractive index, n. Later work[2] verified this treatment in detail and extended the argument to the focal conic texture, which had not been previously recognized as exhibiting reflection colors.[3] In both cases, however, the derived expressions applied only to the special case where the observation direction was in the plane of incidence. The general relationship between angles of incidence, observation, pitch and index of refraction are determined below. Scattering data confirm the expression.

2. Theory

The incident collimated beam is described in terms of the two polar angles θ_i and ϕ_i where θ_i is the angle between the beam and the normal to the air-liquid crystal interface and ϕ_i, the azimuthal angle, is arbitrarily set at zero. The definition of the observation angles θ_s and ϕ_s then follows. The problem is to relate λ, the wavelength, p, the helical pitch,† and n the index of refraction of the cholesteric, to θ_i, θ_s and ϕ_s. It is convenient to introduce two Cartesian coordinate

† The " helical pitch " in this paper corresponds to a 180° turn not the 360° period which was used in Reference 1.

systems, both having an origin at the point of Bragg reflection. The XYZ (unprimed) system is chosen with Z perpendicular to the liquid crystal surface and X in the plane of incidence. The $X'Y'Z'$ (primed) system is chosen such that $Y = Y'$ and Z' is in the direction of the incident beam in the liquid crystal (after refraction). The situation in the XZ plane is shown in Fig. 1, where ψ is the angle

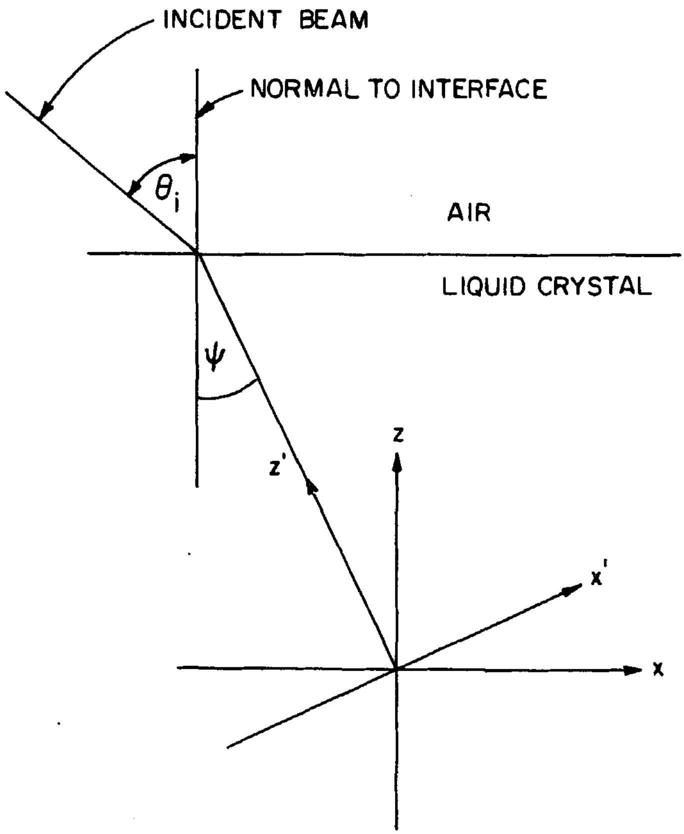

Figure 1. Relationship between primed and unprimed systems.

between the beam and the Z axis in the liquid crystal.

The condition for Bragg reflection is just

$$2np \sin \theta_B = \lambda \tag{1}$$

where θ_B is the Bragg angle and n is the index of refraction. Any Bragg site making this angle with the beam will reflect and the locus of these reflections will be a cone of half angle α. The locus of the normals to the sites is also a cone of $\frac{1}{2}$ the angle of the light cone, as shown in Fig. 2. The effect of refraction for normal incidence ($\theta_i = 0$) is simply to increase the cone angle. A laser beam, for example, produces a ring as has been observed by Durand.[4] Collimated white light produces concentric colored rings. For other

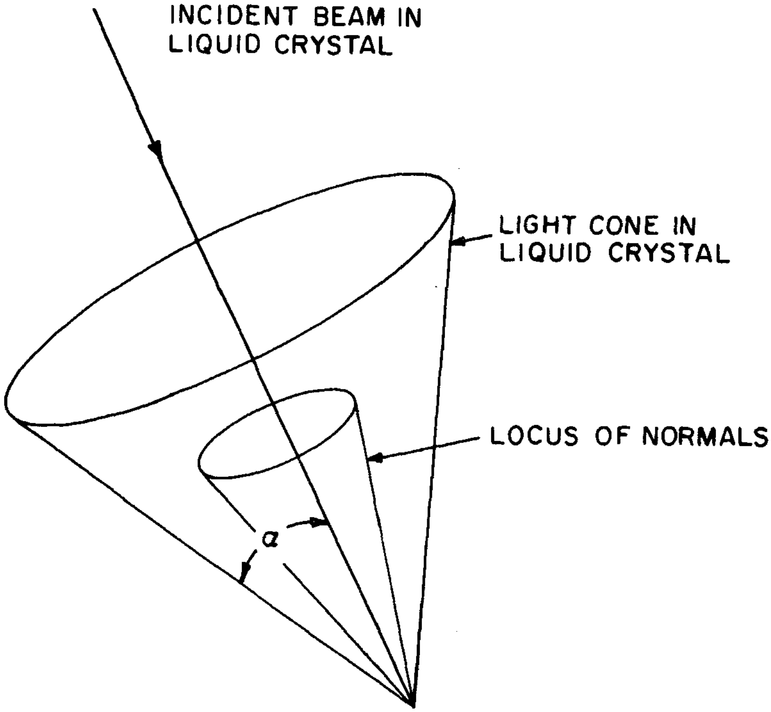

Figure 2. Light cone and cone of normals.

than normal incidence the light cone is distorted by refraction. The equation of the light cone (before refraction) in the primed system is

$$X'^2 + Y'^2 - Z'^2 \tan^2 \alpha = 0 \qquad (2)$$

It is convenient to first transform this equation into the unprimed system giving

$$(X \cos \psi + Z \sin \psi)^2 + Y^2 - \tan^2 \alpha (Z \cos \psi - X \sin \psi)^2 = 0 \quad (3)$$

and finally into a spherical coordinate system with origin identical to the unprimed system. In this system $|\bar{r}|$ is the length of a vector $\bar{r}$ from the origin to a point of interest. θ is the angle between $\bar{r}$ and Z and ϕ is the angle between the projection of $\bar{r}$ in the XY plane and the X axis. In the spherical coordinate system the light cone is described by

$$(\sin\theta\cos\phi\cos\psi + \cos\theta\sin\psi)^2 + (\sin\theta\sin\phi)^2 - \tan^2\alpha(\cos\theta\cos\psi - \sin\theta\cos\phi\sin\psi)^2 = 0 \quad (4)$$

which reduces to ($\sec\alpha \neq 0$)

$$\cos\theta\cos\psi - \sin\theta\cos\phi\sin\psi = \cos\alpha \quad (5)$$

The details of the Bragg reflection are shown in Fig. 3, and evidently

$$\theta_B = (\pi - \alpha)/2 \quad (6)$$

combining Eq. (4), Eq. (5) and Eq. (6) with Snell's law gives

$$\lambda = 2np\cos\tfrac{1}{2}[\cos^{-1} \cdot \{1/n^2(\sqrt{(n^2 - \sin^2\theta_i)(n^2 - \sin^2\theta_s)} - \cos\phi_s\sin\theta_s\sin\theta_i)\}] \quad (7)$$

This is the general relationship among the experimental observables

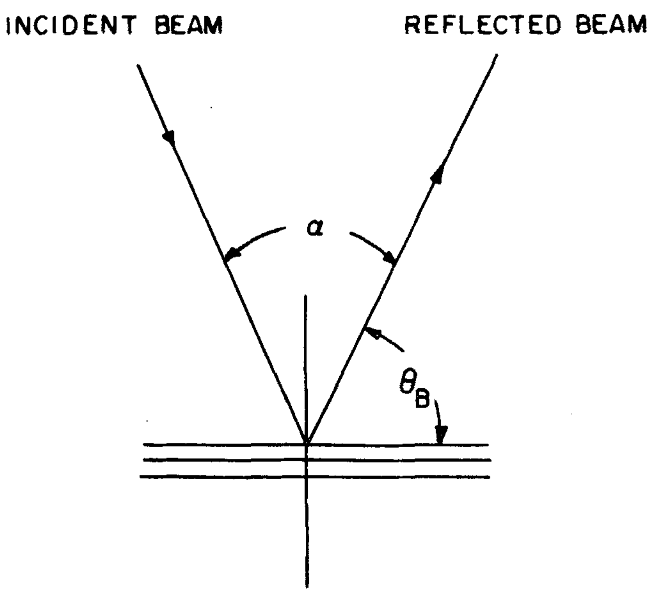

Figure 3. Details of Bragg reflection.

and film parameters. There is no restriction on θ except that

$$\pi/2 + \sin^{-1}\left(\frac{1}{n}\right) \geqslant \theta \geqslant \sin^{-1}\left(\frac{1}{n}\right)$$

will result in total internal reflection. When $0 < \theta < \sin^{-1}(1/n)$, $\theta < \theta_s < \pi/2$ and the Bragg reflected beam will emerge on the same side of the film as the incident beam. When $\pi/2 + \sin^{-1}(1/n) < \theta < \pi$ the reflected beam will appear on the opposite side. For a given λ it is possible that part of the cone emerges on one side of the film and part on the other. For this to occur it is necessary that the critical angle is greater than 45° or that $n < \sqrt{2}$.

So far no assumption was made about the orientational distribution of sites. This distribution, which depends strongly on texture,[5] will affect only the intensity of reflections. For example, for normal incidence, the Grandjean texture will produce the most intense rings for $\theta_s \approx 0$, whereas the focal conic texture will produce the most intense rings for $\theta_s \approx \pi$. In addition to the refraction of the light cone at the liquid crystal-air interface, there will be a partial reflection, the magnitude of which depends on relative indices of refraction and, consequently, for every cone, there will be a second cone caused by reflection. The equation for the reflected cone is given by substituting $(\pi - \theta_i)$ for θ_i in Eq. (7). This applies for all values of θ_i. A glass substrate and/or cover slip will not affect the reflection (there will be a small reflection at any liquid crystal-glass interface but the indexes are so close the intensity will be small).

3. Experimental Procedure

A mixture of 31% by weight cholesteryl nonanoate and 69% by weight cholesteryl chloride was used to provide a convenient wavelength range and sample stability. This particular mixture has the added experimental advantage that its pitch is very insensitive to temperature variations around room temperature.[6] Chemicals were purchased from Eastman Kodak and purified by recrystallization from ethyl alcohol. Ingredients were heated together above the isotropic point and mixed thoroughly. Samples were deposited on a black substrate and were caused to adopt the Grandjean texture. All data were taken at room temperature. The experimental arrangement is shown in Fig. 3. The stage consists of an aluminum block with a sloping face making an angle Σ with the normal to the plane defined by the incident and observed beams. The liquid crystal film is on the sloping face. In this particular experiment the wavelength of incident light (produced by a monochromator) and angle of incidence were fixed and a spatial scan determined the angle maximum reflectance. The reflected signal is not completely symmetric for a variety of reasons[2] and, therefore, the peak in the broadened signal will not correspond exactly to the predicted angles in Eq. (7). In general, if the functional form of the signal is known and the functional forms of all broadening mechanisms are known it is possible to adjust for this effect[7] but this is beyond the scope of this work. In this experiment, the signal widths are sufficiently small that the error introduced is less than $\frac{1}{4}°$. A discussion of other errors intrinsic to this experimental technique is contained in Reference 2.

The experimentally measured angles are A and B as shown in Fig. 4. A is the angle between the incident beam and Z^* where Z^* is in the plane defined by the incident and observed beam and Z^* is perpendicular to Y^* where Y^* is in the XY plane (plane of the

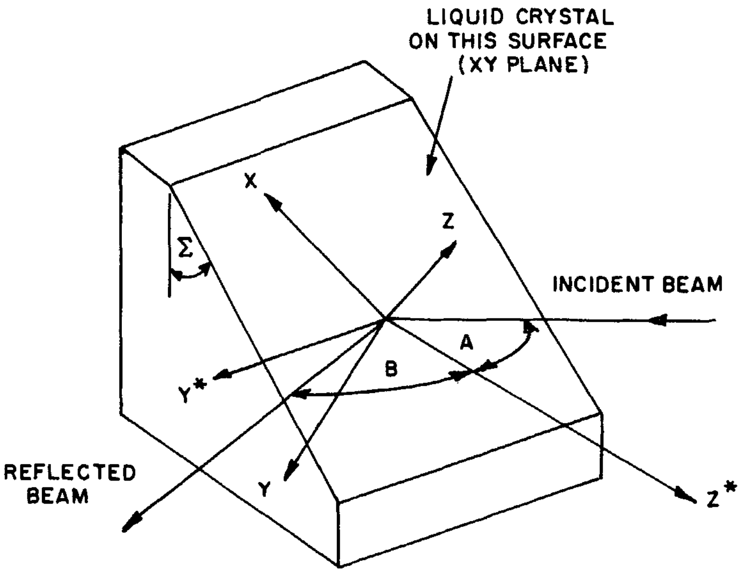

Figure 4. Experimental geometry.

sample). To verify Eq. (7) it is necessary to express θ_i, θ_s, and ϕ_s in terms of A, B and Σ. This is accomplished by first transforming X^*, Y^* and Z^* into an intermediate system X^+, Y^+, Z^+ by rotating around Y^* through an angle Σ and then rotating around Z^+ through an angle Φ ending in the XYZ system. The relationship between the X^* Y^* Z^* and the XYZ system is given by

$$\begin{pmatrix} X \\ Y \\ Z \end{pmatrix} = \begin{pmatrix} \cos\Phi\cos\Sigma & \sin\Phi & -\cos\Phi\sin\Sigma \\ -\sin\Phi\cos\Sigma & \cos\Phi & \sin\Phi\sin\Sigma \\ \sin\Sigma & 0 & \cos\Sigma \end{pmatrix} \begin{pmatrix} X^* \\ Y^* \\ Z^* \end{pmatrix} \tag{8}$$

and Φ is found by insisting that the incident beam have azimuthal angle 0 in the XYZ system yielding

$$\tan\Phi = \tan A \csc\Sigma \tag{9}$$

by combining (7) and (8) the relationships between A, B and Σ and θ_i, θ_s and ϕ_s are found to be

$$\begin{aligned} \cos\theta_i &= \cos A\cos\Sigma \\ \cos\theta_s &= \cos B\cos\Sigma \\ \tan\phi_s &= \frac{\tan A + \tan B}{\tan A\tan B\csc\Sigma - \sin\Sigma}. \end{aligned} \tag{10}$$

4. Experimental Results

Data was taken for stage angles $\Sigma = 10°, 20°, 30°, 40°, 50°$ and $60°$ for a wide range of A and B. About 70 combinations of A and B were measured for each Σ spanning as broad a range as was experimentally possible. The data were computer fitted to Eq. (7) leaving n and p as parameters to be found. The results for $\Sigma = 60°$ are shown in Fig. 5. The solid lines represent the computer calculated best fit to the entire data set. The index of refraction was determined to be 1.49, and p was determined to be 2120 Å. These data compare favorably with an independent measurement based on transmission data from a Cary spectrometer, which give a value of $2np$ of 6280 Å.

5. Remarks

It is recognized that the assignment of a single index or refraction

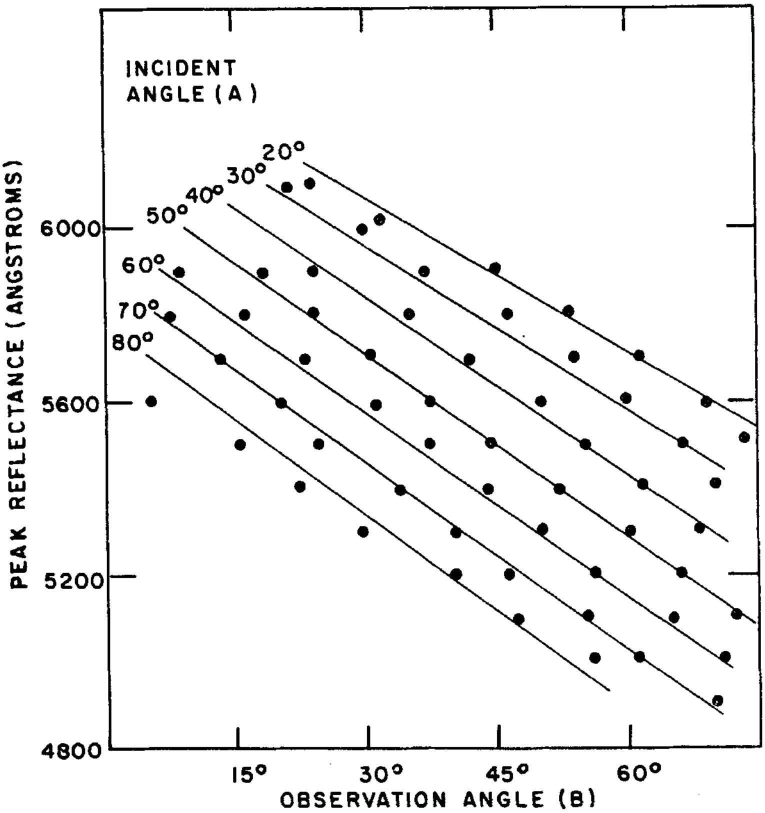

Figure 5. Experimental results for $\Sigma = 60°$. Solid lines represent a computer fit to Eq. 7.

to the cholesteric film is an approximation. In the case of the Grandjean texture, the refractive properties are correctly described by a single optical indicatrix which is an ellipsoid of revolution. Formally, Eq. (7) should be adjusted to include all the complications of double refraction, etc. However, the birefringence is relatively small and its effects are correspondingly small. A complete analysis involving these effects leads to a cumbersome and not particularly useful expression. In the case of the focal conic texture the situation is even more complicated since each helical domain has an indicatrix with an orientation different from its neighbours. Assigning an average index of refraction to these films provides a convenient approximation resulting in a usable expression.

In summary, an expression has been developed which relates the pitch and effective index of refraction to angles of illumination and scattering in cholesteric films and it is applicable to both the Grandjean and focal conic textures. The expression was verified experimentally for a wide range of illumination and viewing conditions.

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