

On the Determination of the Cholesteric Screw Sense by the Grandjean-Cano-Method

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A theoretical treatment is presented for the determination of the screw sense of a cholesteric substance in a Grandjean-Cano-wedge by observing the sample in a polarizing microscope. For comparison some experimental measurements are given.

1. Introduction

A simple and accurate method to determine the pitch of a cholesteric substance is the observation of disclination lines in a Grandjean-Cano-wedge [1–3]. However, from the position of these lines nothing can be said about the sense of rotation of the cholesteric helix. The determination of the sense is often made by either an additional experiment [4] or by a special construction of the wedge allowing to rotate its boundaries [5]. Berthault et al. [6] give a rule which allows to determine the screw sense directly by observing a Grandjean-Cano-wedge under a polarizing microscope. Their argumentation is however very scetchy and the condition of applicability is unclear. For this reason we present here a coherent derivation of the rule.

2. Experimental Geometry and Rule to Determine Sense of Rotation

The geometry of the experiment is shown in Figure 1. The cholesteric layer is placed in the x - y -plane with the director aligned parallel to the x -axis at both boundaries. Light propagating parallel to the z -axis being polarized along the x -direction is used. The sample is observed through an analyser. When polarizer and analyser are crossed one can see parallel to the disclination lines a system of interference stripes. The latter are most pronounced at small wedge thickness. When the analyser is rotated the stripes shift along the wedge gradient. This well-known phenomenon has been used by Stumpf to determine the optical rotatory power of cholesteric samples [7]. It also allows to determine the sense

of rotation of the cholesteric helix according to the following rule [6]:

1) Rotate the analyser in that sense which induces a shift of the interference pattern to regions of smaller wedge-thickness.

2) Combine the rotational motion of the analyser with a translation along the direction of light propagation to obtain the sense of rotation of the cholesteric helix.

3) This rule holds when the wavelength λ of the light is small compared to the pitch P or more accurately for

$$\lambda < P n_0 n_e (n_e^2 + n_0^2)^{-1/2}, \quad (2.1)$$

where n_0 and n_e are ordinary and extraordinary indices of refraction. This condition is usually fulfilled when the Grandjean-Cano-wedge method can be applied.

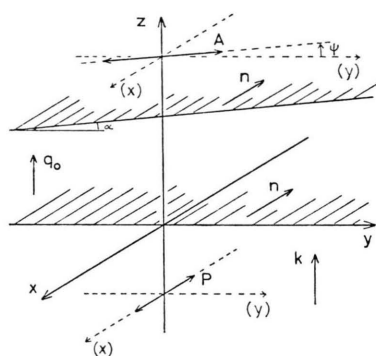


Fig. 1. Coordinate system with the wedge, the directors \mathbf{n} at the surfaces and Polarizer P and Analyser A which is rotated by the angle ψ from the crossed position. Incident light (k) is from the bottom. The wave vector q_0 of the cholesteric screw is essentially along the z -direction since the wedge angle α is almost zero.

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3. Theoretical Aspects

In the sample the director \mathbf{n} has the form:

$$\mathbf{n} = (\cos q_0 z, \sin q_0 z, 0), \quad (3.1)$$

where a positive value of q_0 corresponds to a right-handed helix in the coordinate system chosen in Figure 1.

The electrical field vector

$$\mathbf{E}(z, t) = \text{Re} \{ \mathbf{E}(z) e^{-i\omega t} \}$$

of a wave travelling through the sample in z -direction is assumed to be perfectly linearly polarized along the x -direction before entering the sample; i. e. $E_y(z=0) = 0$, $E_x(z=0) = E$. Then at the distance $z = m \pi / |q_0|$ (which is $m/2$ times the value of the pitch P) the electric field is [3]

$$\begin{aligned} E_x(m \pi / q_0) &= \frac{(-1)^m E}{\text{ctg } \varphi_2 - \text{ctg } \varphi_1} \\ &\quad \cdot \{ \text{ctg } \varphi_1 e^{i\delta_1 m} - \text{ctg } \varphi_2 e^{i\delta_2 m} \} \\ E_y(m \pi / q_0) &= \frac{i(-1)^m E}{\text{ctg } \varphi_2 - \text{ctg } \varphi_1} \{ e^{i\delta_1 m} - e^{i\delta_2 m} \}. \end{aligned} \quad (3.2)$$

the phases $\delta_j = \pi l_j / |q_0|$ contain the wave-vectors l_j which are the solutions of the dispersion relation [3]:

$$l_j^2 = k_0^2 + q_0^2 \pm \sqrt{4 q_0^2 k_0^2 + k_1^4}, \quad (3.3)$$

where

$$\begin{aligned} k_0^2 &= \frac{1}{2} (\omega/c)^2 (n_e^2 + n_o^2), \\ k_1^2 &= \frac{1}{2} (\omega/c)^2 (n_e^2 - n_o^2). \end{aligned} \quad (3.4)$$

For the moment we consider positive optical anisotropies ($n_e > n_o$ or $k_1^2 > 0$). But this is no restriction as will be shown later on. The angles φ_j are given [3] by:

$$\text{tg} \left(\varphi_j + \frac{\pi}{4} \right) = \frac{(l_j + q_0)^2 - k_0^2}{k_1^2} = \frac{k_1^2}{(l_j - q_0)^2 - k_0^2} \quad (3.5)$$

For our purpose it is sufficient to consider frequencies above the Bragg reflection gap which is equivalent to the condition [3]

$$k_0^2 - k_1^2 > q_0^2. \quad (3.6)$$

Then for a wave travelling in positive z -direction the positive values for the wave vectors l_j , corresponding to positive group velocities have to be taken [3]. At the upper plate of the wedge the director has made a rotation by an integer multiple m_0 of π according to the given boundary conditions

on the sample. If the analyser is rotated by a small angle ψ from the y -direction (see Fig. 1), the electric field of the observed light is given by

$$\begin{aligned} E_{\text{obs}} &= E_y \left(m_0 \frac{\pi}{q_0} \right) \cdot \cos \psi - E_x \left(m_0 \frac{\pi}{q_0} \right) \sin \psi \\ &\approx \frac{i(-1)^{m_0} E}{\text{ctg } \varphi_2 - \text{ctg } \varphi_1} \{ e^{i(\delta_1 m_0 + \psi \text{ctg } \varphi_1)} - e^{i(\delta_2 m_0 + \psi \text{ctg } \varphi_2)} \} \end{aligned} \quad (3.7)$$

as follows from (3.2) and the approximation

$$\cos \psi + i \sin \psi \text{ctg } \varphi_i \approx \exp \{ i \psi \text{ctg } \varphi_i \}$$

which is valid to order ψ .

The observed interference pattern and its dependence on the angle ψ is essentially determined by the phase difference Δ in (3.7), where

$$\Delta = (\delta_1 - \delta_2) m_0 + \psi (\text{ctg } \varphi_1 - \text{ctg } \varphi_2). \quad (3.8)$$

In order to understand the dependence of the interference pattern on ψ we have to examine the quantities $\text{ctg } \varphi_j$ which, from (3.5), read

$$\begin{aligned} \text{ctg } \varphi_j &= \frac{(l_j + q_0)^2 - k_0^2 + k_1^2}{(l_j + q_0)^2 - k_0^2 - k_1^2} \\ &= \frac{(l_j - q_0)^2 - k_0^2 + k_1^2}{-(l_j - q_0)^2 + k_0^2 + k_1^2}. \end{aligned} \quad (3.9)$$

First we consider the case $q_0 > 0$, $k_1^2 > 0$. Then replacing (3.3) for l^2 in (3.9) yields

$$\text{ctg } \varphi_1 = \frac{2(l_1 + q_0)q_0 + \sqrt{4q_0^2 k_0^2 + k_1^4} + k_1^2}{2(l_1 + q_0)q_0 + \sqrt{4q_0^2 k_0^2 + k_1^4} - k_1^2} > 1, \quad (3.10)$$

which is obviously greater than one. Similarly we can write

$$\text{ctg } \varphi_2 = - \frac{2l_2 q_0 - 2q_0^2 + \sqrt{4q_0^2 k_0^2 + k_1^4} - k_1^2}{2l_2 q_0 - 2q_0^2 + \sqrt{4q_0^2 k_0^2 + k_1^4} + k_1^2}. \quad (3.11)$$

From the relation

$$\sqrt{4q_0^2 k_0^2 + k_1^4} > 2q_0^2 + k_1^2, \quad (3.12)$$

which follows from (3.6), we obtain

$$-1 < \text{ctg } \varphi_2 < 0. \quad (3.13)$$

Together with (3.10) this leads to the result

$$\text{ctg } \varphi_1 - \text{ctg } \varphi_2 > 1, \quad q_0 > 0. \quad (3.14)$$

For negative optical anisotropy, i. e. $k_1^2 < 0$ or $n_e < n_o$ (which is also equivalent to polarizing the entering light along the y -direction in the positive case) it follows from (3.9) that $\text{ctg } \varphi_i$ have the

reciprocal values of the corresponding positive case. Thus the relation (3.14) remains unchanged.

Considering now a lefthanded cholesteric where $q_0 < 0$ we see from (3.9) that $\text{ctg } \varphi_i$ changes its sign whereas its absolute value does not change. This leads to the following modification of relation (3.14):

$$\text{ctg } \varphi_1 - \text{ctg } \varphi_2 < -1, \quad q_0 < 0. \quad (3.15)$$

From (3.14) and (3.15) follows now how the phase difference Δ changes in (3.8) when the analyser angle ψ is altered. In order to understand the resulting movement of the interference fringes the change of Δ with changing $|q_0|$ is calculated (between two dislocation lines a change in the wedge thickness z_0 corresponds to a change in q_0 because $z_0 = m_0 \pi / |q_0|$). From

$$\frac{d\Delta}{dz_0} = \frac{d}{d|q_0^{-1}|} \left[\frac{l_1}{|q_0|} - \frac{l_2}{|q_0|} \right] \quad (3.16)$$

follows the necessity to examine the quantities

$$D_j = \frac{d}{d|q_0^{-1}|} \frac{l_j}{|q_0|} \equiv |l_j|^{-1} \frac{d}{d(q_0^{-2})} \left(\frac{l_j}{q_0} \right)^2 \quad (3.17)$$

$$= \left[\frac{k_0^2}{q_0^2} \pm \left(2 \frac{k_0^2}{q_0^2} + \frac{k_1^4}{q_0^4} \right) \left(4 \frac{k_0^2}{q_0^2} + \frac{k_1^4}{q_0^4} \right)^{-1/2} \right] \cdot \frac{q_0^2}{l_j}.$$

In particular we want to know under what condition $d\Delta/dz_0$ is positive which implies

$$D_1 > D_2. \quad (3.18)$$

After some rearrangements one arrives at the result that this relation is equivalent to

$$k_0^4 - k_1^4 > 2 k_0^2 q_0^2. \quad (3.19)$$

Inserting for k_0^2 and k_1^2 from (3.4) we obtain at a given wavelength $\lambda = 2\pi c/\omega$ the condition for positive $d\Delta/dz_0$

$$\lambda/P < n_e n_0 / \sqrt{n_e^2 + n_0^2}. \quad (3.20)$$

If the movement of interference fringes is observed the phase difference Δ is kept constant hence one obtains

$$\frac{dz_0}{d\psi} \equiv - \frac{(\partial\Delta/\partial\psi)_{z_0}}{(\partial\Delta/\partial z_0)_\psi}. \quad (3.21)$$

Combining all the results we may state: For a righthanded cholesteric ($q_0 > 0$) the interference fringes move towards regions of smaller wedge thickness z_0 when the analyser angle ψ increases provided condition (3.20) is satisfied. For a left-

handed cholesteric a movement in the opposite direction occurs: This is the rule stated in Section 2.

The above treatment can be generalized in that the incoming light may be polarized in a direction rotated by an angle χ with respect to the x -axis (positive χ in the same sense as pos. ψ). Then the relevant phase difference for the interference pattern has the form

$$\Delta = (\delta_1 - \delta_2) m_0 + \arctg \left(\frac{\text{tg } \chi}{\text{tg } \varphi_2} \right) - \arctg \left(\frac{\text{tg } \chi}{\text{tg } \varphi_1} \right) \\ + \arctg \left(\frac{\text{tg } \psi}{\text{tg } \varphi_1} \right) - \arctg \left(\frac{\text{tg } \psi}{\text{tg } \varphi_2} \right). \quad (3.22)$$

From this expression it follows that the stated rule remains true for any value of χ . Furthermore since Δ increases monotonically with ψ (see 3.10 and 3.13) the rule is not restricted to small values of ψ . However, the interference pattern is most pronounced for the geometry chosen (and for $\chi = \pi/2$, $\psi = \pi/2$) because for other angles there is generally a constant background which may be considerably more intensive than the interference modulation.

It is of course possible to formulate an analogous rule for the case of rotating the polarizer instead of the analyser. This rule is easily derived from Equation (3.22).

Clearly our special choice of boundary conditions is no restriction either. For other homogeneous directions of alignment only the angles ψ and χ have to be defined with respect to the new directions.

4. Experimental Example

In order to test the theoretical arguments of the last section we have measured the shift of the interference pattern as observed in two cholesteric samples. They are mixtures of the nematic mixture RO-TN-403 from F. Hoffmann-La Roche [8] with the cholesteric dopands 4-Cyano-4'-(2-methylbutyl)-biphenyl (CB 15 from BDH) and Cholesterylchlorid (CC from Eastman) which are known to induce right- and lefthanded cholesteric helixes respectively when added in low concentration to similar nematic mixtures [5]. Mixture C1 contained 2.21 wt% of CB15 in RO-TN-403 while C2 contained 4.98 wt% of CC in RO-TN-403.

These substances were filled into a wedge whose thickness varied by 50 microns over its length of about 27 mm. Both surfaces were treated by 30°

oblique evaporation of SiO in such a way that the nematic director pointed normal to the thickness gradient (Figure 1). The whole wedge which was placed under a polarizing microscope could be moved in the direction of the thickness gradient by means of a micrometer screw. The linearly polarized light was filtered through a 497 nm interference filter of 40 nm width. The pitch of the two mixtures at room temperature was 6.3μ for C1 and 6.1μ for C2. We have measured the position of an interference minimum as a function of the angle ψ (Fig. 1) for $-20^\circ < \psi < 20^\circ$. With the known wedge geometry we could determine the wedge thickness at a given position.

Figure 2 shows a plot of the thickness difference $z_0(\psi) - z_0(\psi=0)$ at the position of minimal light intensity as a function of the angle ψ . From the polynomial fits drawn in Fig. 2 we obtain the derivatives

$$\left. \frac{dz_0}{d\psi} \right|_{\psi=0} = \begin{cases} -1.19 \mu, & \text{C1 exp,} \\ 1.12 \mu, & \text{C2 exp.} \end{cases} \quad (4.1)$$

This is to be compared with the values calculated from expression (3.21). We have used in the calculation the properties of undoped RO-TN-403 in particular $n_e = 1.7810$ and $n_o = 1.5235$ [8]. With these values we obtain

$$\left. \frac{dz_0}{d\psi} \right|_{\psi=0} = \begin{cases} -1.28 \mu, & \text{C1 calc,} \\ 1.26 \mu, & \text{C2 calc.} \end{cases} \quad (4.2)$$

In view of the fact that the geometry is not perfectly well known (unevenness of glass plates, unaccuracy of spacers thickness), that the refraction indices may be altered somewhat by the dopants and that it is quite difficult to get accurate values for derivatives of measured curves the agreement is satisfactory indeed.

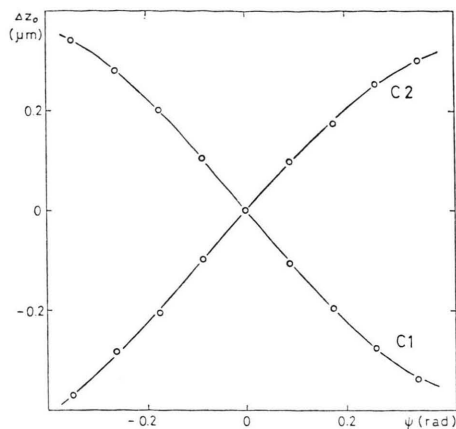


Fig. 2. Wedge height at the position of the interference minimum of green light (497 nm) as a function of the analyser angle ψ_1 . The lines are polynomial fits to the experimental points. The scale is shifted to give zero at $\psi = 0$. For C1 the height decreases with increasing ψ giving a right-handed screw sense. The sample C2 has a left-handed sense. The dissymmetry of the curves may stem from alignment deviations of the directors at the surfaces.

5. Conclusion

We have checked a simple rule for determining the screw sense of a cholesteric liquid. Although the justification of this rule is fairly elaborate, the rule is simple and straight-forward to be applied in routine measurements. Since the Grandjean-Cano-method is well suited for accurate pitch measurements especially for large pitch values, this rule may prove to be useful in many cases.

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