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Pitch Dependence on Strain in Cholesteric Films

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A cholesteric film in the Grandjean texture is placed on an elastic substrate. As a result of a positive strain, the film undergoes a temporary blue shift. The effect can be explained in terms of a direct geometrical argument; i.e., the total angle through which the director rotates is temporarily preserved and because the strained film is thinner, the pitch decreases. The sample relaxes to its original pitch in a few seconds.

INTRODUCTION

The local symmetry of the cholesteric mesophase, at least in the Grandjean and focal-conic textures, is helical.[1] It is characterized by a pitch which corresponds to a 180° rotation of the director. The pitch is sensitive to a variety of stimuli which include temperature changes,[2] electric and magnetic fields,[3] exposure to vapor,[4] and ultraviolet light.[5] In some cases actual compositional changes are involved (irreversible) while in other cases the pitch returns to its original value once the stimulus is removed. When the cholesteric material consists of a mixture of molecular species, the sensitivity of a film can be predicted in terms of the individual sensitivities of its constituents.[6]

The purpose of this communication is to present data relating to another pitch-altering effect and a straightforward interpretation of these data. The stimulus involves subjecting a cholesteric film to a rapid and substantial strain. It is known that when a cholesteric film in the Grandjean texture is placed on a relaxed elastic substrate which is subsequently stretched, the pitch suffers a temporary shift towards the blue. This has been attributed to a decrease in entropy of the substrate due to an increase in orientational order imposed by the stretching and a corresponding increase in entropy (adiabatic process) in the liquid crystal, resulting in a temperature rise and pitch shift.[7] We will show that this is not the dominant mechanism, but rather that the total angle through which the director rotates temporarily remains the same after strain and the thinner film requires a pitch shift towards the blue.

EXPERIMENTAL DETAILS

To obviate thermal arguments, a film was chosen which shifts red with increasing temperature. The material is a mixture of 30% by weight cholesteryl chloride, 10% cholesteryl nonanoate, and 60% oleyl cholesteryl carbonate. A plot of pitch versus temperature is shown in Figure 1. The data were taken using a Cary 14 Spectrometer and a Mettler FP2 to control

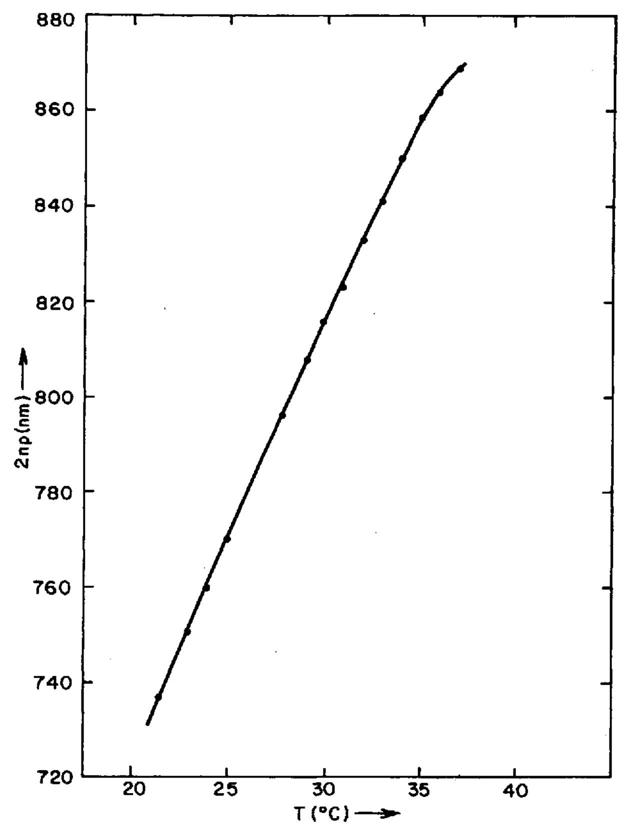

FIGURE 1 Pitch vs temperature—30% by weight cholesteryl chloride, 10% cholesteryl nonanoate and 60% oleyl cholesteryl carbonate.

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temperature. Detailed results are presented for this composition; however, the effect was observed with many compositions and is independent (to within the accuracy of our experiments of the sign and magnitude of dependence on temperature.

The sample was placed on a thin, transparent, elastic substrate which could be strained repeatedly without fatigue (this requirement limited strains to around 1.0). The substrate was chosen such that the film adhered to it well and after several flexings still exhibited continuity. The pitch shift reached its peak very rapidly after expansion (less than a second) and remained in that condition for times of the order of a few seconds, depending on viscosity, etc. The film was mounted vertically. The experimental apparatus is shown in Figure 2. The strain was determined by adjusting the stop and when the latch was withdrawn, the springs would exert a force, causing the substrate and sample to elongate. The strained sample exhibited good Grandjean characteristics with only a little focal-conic clouding. However, when the substrate was returned to its original dimension, the sample converted to a substantially focal-conic condition. Therefore, at the beginning of each measurement, the sample was sheared to ensure a Grandjean starting texture.

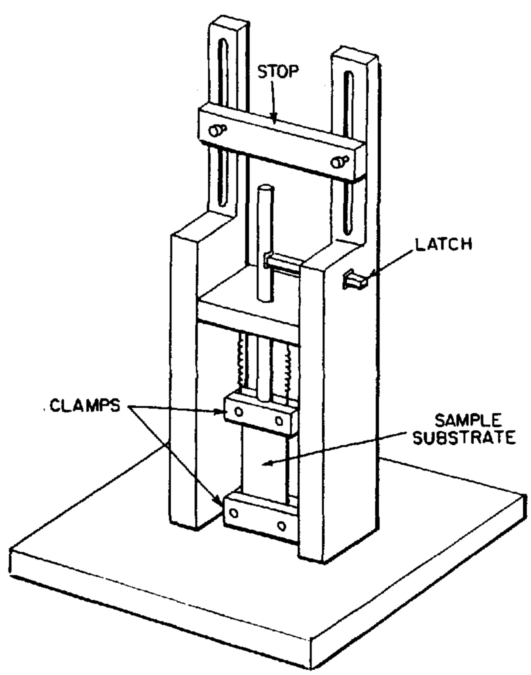

FIGURE 2 Apparatus.

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The pitch was measured in transmission using a Bausch & Lomb monochromator as a source and a photodiode as a detector. For normally incident unpolarized or plane-polarized light, the Grandjean texture reflects approximately 50% of incident light in a wavelength band centered around λ where $\lambda = 2np$ (n is the average index of refraction and p is the pitch).[8] Away from this band, light is essentially completely transmitted, limited of course by absorption in the ultraviolet and infrared. It is therefore possible to determine pitch by determining the wavelength of minimum transmission. Initially, the wavelength $\lambda_0 = 2np_0$, where p_0 is the pitch of the unstrained material, was determined by scanning the wavelength of the incident light until a minimum in transmission was found. Simultaneously with substrate elongation, the monochromator wavelength was quickly decreased from λ_0 until a new minimum in transmission was detected at $\lambda = 2np$, where p is the pitch of the strained material. The determination of λ was repeated several times at each strain setting of the apparatus. With this technique, the measured spread in λ was approximately 30 nm.

The actual strain was calculated based on the geometry shown in Figure 3. This is an indication of the change of shape of the substrate. The liquid

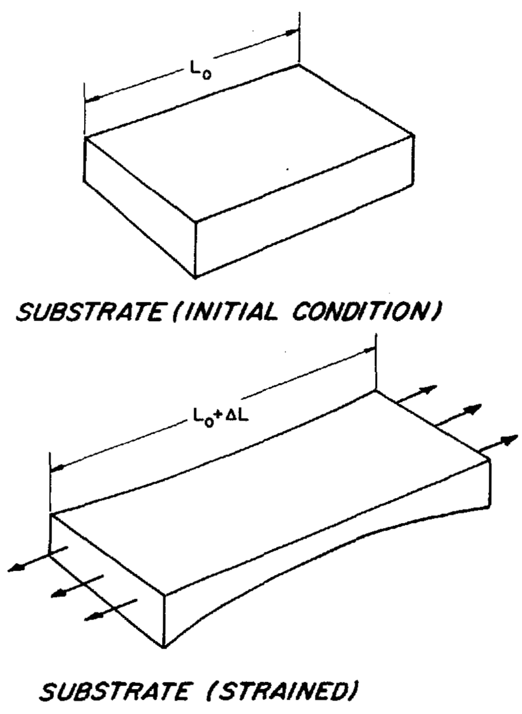

FIGURE 3 Unstrained and strained substrate.

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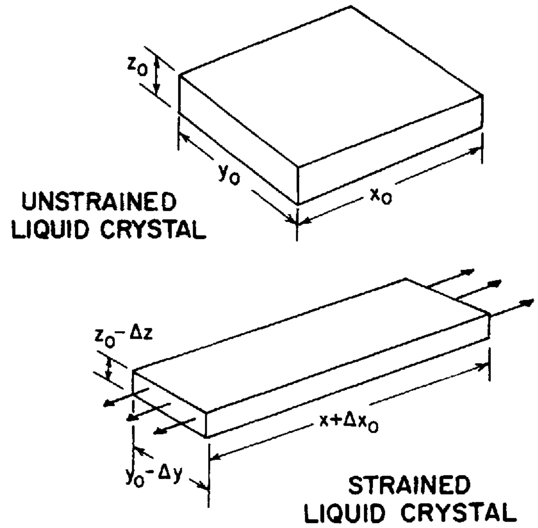

FIGURE 4 Unstrained and strained liquid crystal.

crystal was placed in the center of the substrate to minimize distortions and is assumed to be represented by Figure 4. The change in the liquid-crystal X and Y dimensions was determined by placing fiducial marks on the substrate and measuring the change in their spacing when the sample was elongated in the X direction. It was found empirically that

$$\Delta x/x_0 = \Delta L/L_0 \quad \text{and} \quad \Delta y/y_0 = 0.35\,\Delta L/L_0 \tag{1}$$

where $\Delta L/L_0$ is the strain induced in the substrate.

MODEL

We propose, as a straightforward model, that immediately after strain the total angle through which the director rotates is preserved. Therefore,

$$(\lambda - \lambda_0)/\lambda_0 = (p - p_0)/p_0 = -\Delta z/z_0. \tag{2}$$

Assuming constant volume of the liquid crystal, one can write

$$x_0 y_0 z_0 = (x_0 + \Delta x)(y_0 - \Delta y)(z_0 - \Delta z). \tag{3}$$

By combining Equations (1), (2), and (3) the instantaneous pitch of the strained material is given by

$$p = p_0\left[\left(1 + \frac{\Delta L}{L_0}\right)\left(1 - 0.35\frac{\Delta L}{L_0}\right)\right]^{-1} \tag{4}$$

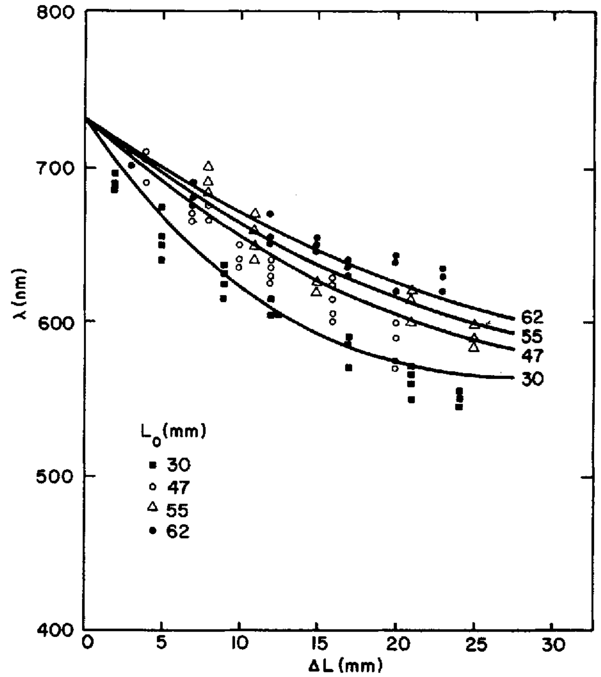

FIGURE 5 Pitch vs strain.

The experimental results for four different values of L_0 are shown in Figure 5. The solid lines are generated by Eq. (4). Similar results were obtained for other combinations of p_0, L_0, ΔL, $\partial p/\partial T$ (T = temperature), etc. It is interesting to observe that the right-hand side of Eq. (4) contains only experimentally fixed variables and is consequently totally unforgiving regarding a good fit.

CONCLUSION

Although thermal effects are certainly present in such an experiment, geometrical considerations dominate. The total director twist is momentarily conserved and a competition between the appropriate elastic constant(s) and viscosities determines the relaxation rate.

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