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# Wavelength Modulation Using Cholesteric Liquid Crystals

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In this paper we report the results of an extended experimental study on the optical rotatory power of Cholesteric Liquid Crystals submitted to compression or dilation. The theoretical background is given by the De Vries' theory modified to take into account the mechanical effects. The experimental data are in excellent agreement with this theory and show variations of the rotatory power of the liquid crystal vs. the applied normal strain with a strong dependence on the light wavelength. Data are reported on the application of this effect to optical modulation. A device operating with these characteristics has been patented.

## INTRODUCTION

The effects of external excitations applied to cholesteric liquid crystals have been studied by us for several years.<sup>1</sup> By detecting the variations observed on the optical properties we have shown that reversible and non-reversible deformations occur when the cholesteric structure is submitted to a static electric field.<sup>2</sup> Moreover we have studied the optoelastic behavior of cholesterics both in the steady state<sup>3</sup> and in the transient regime<sup>4</sup> showing that small normal strains produce elastic deformations.

In this paper we report a study of the effect of these deformations on the rotatory power of Cholesteric Liquid Crystals (C.L.C.) with a generalization of the electromagnetic theory of wave propagation along the helical axis of cholesterics in order to include the case of small mechanical stresses.

The measurements of optical rotatory dispersion under different stress conditions are in excellent agreement with the theoretical model. Data on the variation of the optical rotation vs. the normal strain are reported showing a strong dependence on the light wavelength.

These results and the data on the optical modulation that can be achieved are the basis of the operating characteristics of an optical device recently patented by us: the Stress Controlled Polarizer (SCP).<sup>5</sup> Using this device it is possible to control the polarization of the transmitted light by merely inducing a small compression or dilation to the liquid crystal cell. The optical behavior of the SCP is very similar to that of the VCP (Voltage Controlled Polarizer)<sup>6</sup> described elsewhere and previously patented,<sup>7</sup> but the physical effect which is exploited is completely different.

## THEORETICAL BACKGROUND

For small deformations we have demonstrated<sup>3</sup> a steady state elastic behavior for which the following relation holds:  $\Delta P/P_0 = \delta/d$  where  $P_0$  is the pitch of the unperturbed helix,  $\Delta P$  is the pitch variation when the sample thickness  $d$  is changed by  $\delta$  (dilated if  $\delta > 0$ , or compressed if  $\delta < 0$ ). The small deformations limit means that  $\delta \ll d$ .

In our model the cholesteric structure is compressed or dilated in a direction parallel to the helical axis, so that for small deformations we disregard any rotation or tilt of the molecules with respect to the molecular layer. Therefore we consider just a linear translation of the molecules parallel to the helical axis.

According to this picture, the azimuthal angle describing the local orientation of the director is given by

$$\theta(z) = 2\pi(z - u)/P_0 \quad (1)$$

where  $u \equiv u(z)$  is the displacement of the molecular layer.

In the elastic regime  $u = \alpha z$ , where

$$\alpha = \delta/(d + \delta). \quad (2)$$

For a plane wave travelling along the  $z$ -axis, Maxwell eqs. give

$$\partial^2 \vec{E}_\perp / \partial z^2 + \omega^2 \epsilon \vec{E}_\perp / c^2 = 0 \quad (3)$$

where  $|E_\perp| = \sqrt{E_x^2 + E_y^2}$  and for each term the usual definitions are given. Then we solve the problem in the rotating frame  $(\xi, \eta, z)$ , where

the helix is unwinded and the dielectric tensor has the simple form:<sup>8</sup>

$$\vec{\epsilon}_T \equiv \begin{pmatrix} \epsilon_\xi & 0 & 0 \\ 0 & \epsilon_\eta & 0 \\ 0 & 0 & \epsilon_\eta \end{pmatrix} \quad (4)$$

By applying the operator  $\vec{R} \equiv \begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix}$  we get the new wave equation:

$$[\partial^2/\partial z^2 + 2(\partial\theta/\partial z)\vec{\sigma} \partial/\partial z - (\partial\theta/\partial z)^2 + \omega^2\vec{\epsilon}_T/c^2] \vec{E}_{\perp T} = 0 \quad (5)$$

$$\text{where } \vec{\sigma} \equiv \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}.$$

Looking for a plane wave solution:

$$\vec{E}_T = \sum_j (E_\xi \hat{\xi} + E_\eta \hat{\eta})_j e^{i(k_{jz} - \omega t)} \quad (6)$$

we easily get the dispersion relation

$$\begin{aligned} k^4 + \{-(\omega^2/c^2)(\epsilon_\xi + \epsilon_\eta) - 2(\partial\theta/\partial z)^2\}k^2 \\ + 4i(\partial\theta/\partial z)(\partial^2\theta/\partial z^2)k + (\omega^2/c^2)^2\epsilon_\xi\epsilon_\eta - (\omega^2/c^2)(\epsilon_\xi + \epsilon_\eta)(\partial\theta/\partial z)^2 \\ + (\partial\theta/\partial z)^4 + (\partial^2\theta/\partial z^2)^2 = 0 \end{aligned} \quad (7)$$

Using Eq. (1) in (7) we can write the dispersion relation as:

$$\tilde{K}^4 - 2(1/\lambda_p'^2 + 1)\tilde{K}^2 + \{1 - (\delta e)^2/\lambda_p'^4 - 2/\lambda_p'^2 + 1\} = 0 \quad (8)$$

where the following definitions have been used:

$$\epsilon = (\epsilon_\xi + \epsilon_\eta)/2; \quad \delta\epsilon = (\epsilon_\xi - \epsilon_\eta)/2\epsilon; \quad \lambda' = (2\pi/P_0)(c/\omega) 1/\sqrt{\epsilon};$$

$$P = P_0/(1 - \alpha); \quad \lambda_p' = \lambda'(1 - \alpha); \quad \tilde{K} = KP/2\pi$$

Eq. (8) is the same equation deduced by H. De Vries<sup>9</sup> for the unperturbed cholesteric, taking into account the different definitions of the parameters.

According to that equation, the stressed cholesteric liquid crystal in the elastic regime behaves like the unperturbed one with the new pitch

$$P = P_0/(1 - \alpha)$$

Taking into account this change we can apply to the deformed cholesteric the same expression for the rotatory power

$$R = -(2\pi/P)[(\delta\epsilon)^2/(8\lambda_p'^2(1 - \lambda_p'^2))]. \quad (9)$$

## MEASUREMENTS OF OPTICAL ROTATION

A sketch of the experimental set-up used to perform the measurements of optical rotation is shown in Figure 1. We have used the method of the spinning analyzer in order to improve the accuracy of these measurements. The sample is placed between a fixed polarizer and an analyzer rotating at a constant speed: a square wave signal is detected by the photomultiplier. A reference signal at the same frequency is obtained on the photodiode placed in front of the reflecting

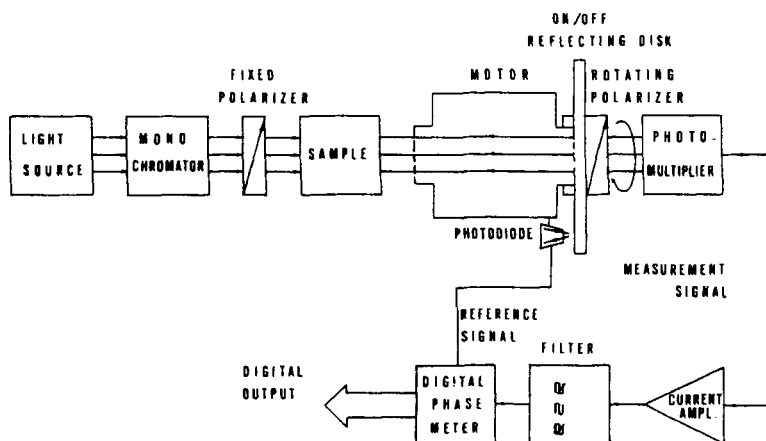


FIGURE 1 Experimental set-up for measurement of optical rotation.

disk rotating at the same speed of the analyzer. The phase delay between these two signals gives exactly two times the optical rotation induced by the liquid crystal sample.<sup>10</sup> A built-in digital phase meter was used to have a direct reading of the optical rotation with an accuracy of  $0.5^\circ$ .

The samples used were mixtures of cholesteryl nonanoate, cholesteryl oleate and cholesteryl chloride placed between two optical glasses with a surface treatment which produces planar alignment of the liquid crystal. The sample holder, which has been described elsewhere,<sup>3</sup> was designed to allow compression or dilation using piezoelectric ceramics glued to one of the two glasses. In Figure 2 we report typical experimental data of the rotatory dispersion curve under different strain conditions for a  $75\text{ }\mu\text{m}$  thick C.L.C. Here optical rotation is reported vs. the light wavelength. In the figure data obtained with no perturbation ( $\delta = 0$ ), small dilation ( $\delta/d = 0.01$ ) and small compression ( $\delta/d = -0.01$ ) are reported. We observe, as expected, an increase of the apparent pitch for the dilated sample, and a decrease for the compressed one. The full lines are calculated from the reported theory. The material parameters have been adjusted to fit the data corresponding to the unperturbed sample. Using the same parameters the data corresponding to dilation and compression have been fitted. We find an excellent agreement between theory and

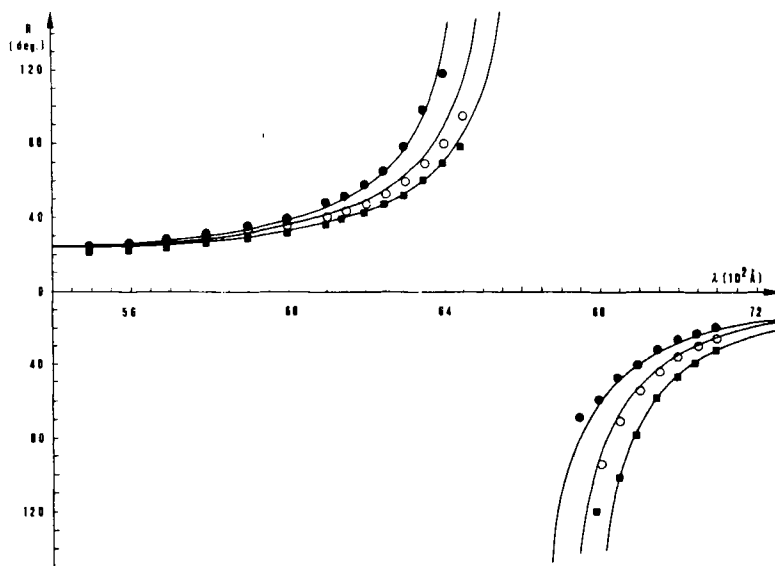


FIGURE 2 Optical rotation vs. light wavelength for different strain conditions. Circles:  $\delta/d = 0$ ; dots:  $\delta/d = 0.01$ ; squares:  $\delta/d = -0.01$ .

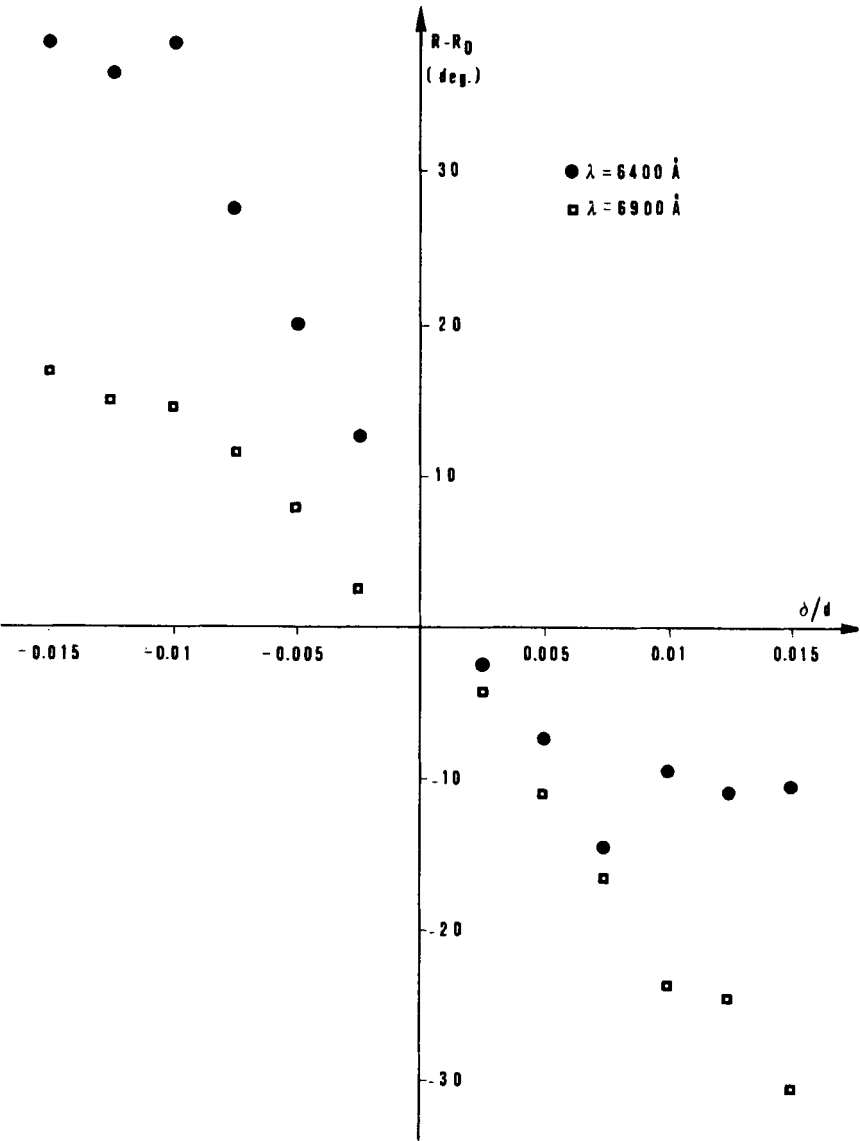


FIGURE 3 Variation of the optical rotation vs. the mechanical strain  $\delta/d$ .  $R_0$  is the optical rotation of the unperturbed sample,  $R$  is the optical rotation under compression or dilation.

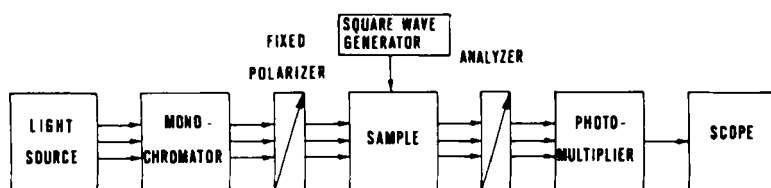


FIGURE 4 Experimental set up for measurement of light modulation.

experimental data. The agreement fails only close to the total reflection region where, of course, the rotatory power is no longer defined in the frame of the present theory.

An important feature to be noted is the strong variation of the optical rotation at a fixed wavelength in the region close to the reflection band, in a way similar to what we have previously reported in the case of an external applied electric field.

For example in Figure 2 we observe that for  $\lambda = 6800 \text{ \AA}$ , the

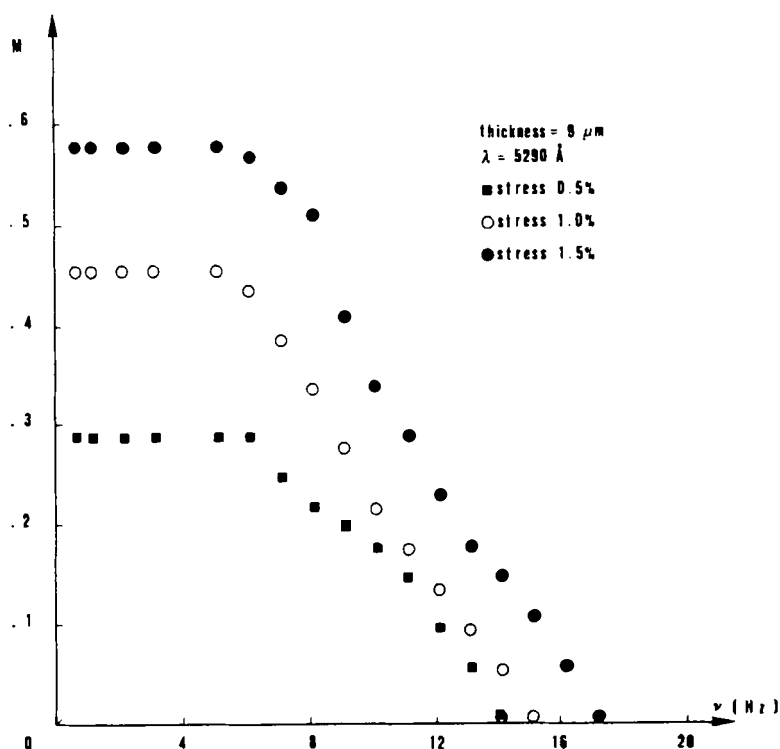


FIGURE 5 Light modulation  $M$  vs. the excitation frequency for a sample  $9 \mu\text{m}$  thick.

optical rotation changes by about  $70^\circ$  going from a small compression to a small dilation.

This is better explained by Figure 3 where the variation of optical rotation is reported vs. the applied strain for different wavelengths for the same sample. There  $R_0$  is the optical rotation of the unperturbed sample and  $R$  is the optical rotation under mechanical stress. The strong dependence of the effect on the light wavelength is well shown in this figure where big variations of optical rotation are reported for wavelengths close to the reflection band.

These data point out the advantage of using for practical applications this elastic effect instead of the electric field induced deformation. In the former case indeed, it is possible to scan through the unperturbed state in both directions, i.e. by increasing or decreasing the rotatory power at a fixed wavelength in order to exploit a wider range of variations.

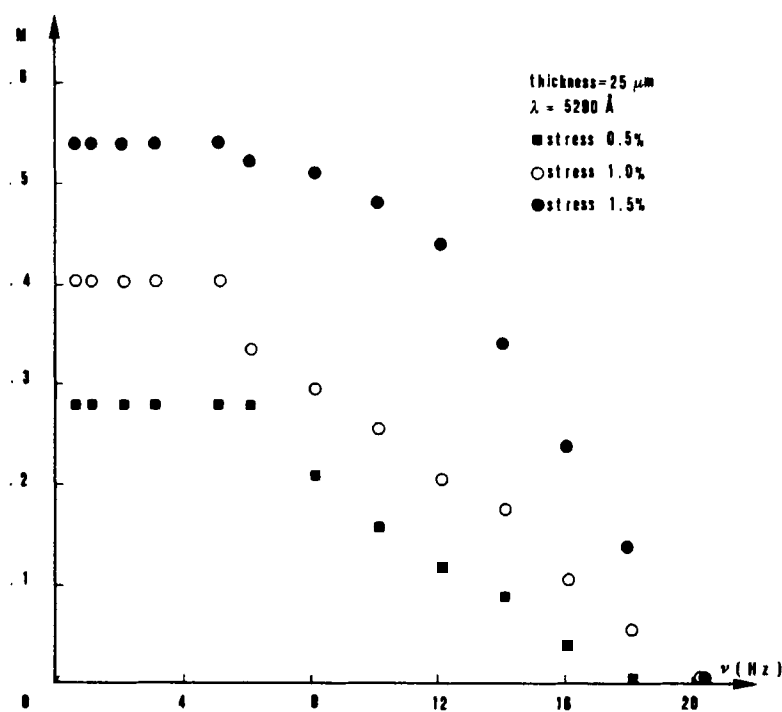


FIGURE 6 Same as Figure 5 for a sample 25  $\mu\text{m}$  thick.

## OPERATING CHARACTERISTICS OF THE SCP

The effects described in the former section are the basis for the working operation of a Stress Controlled Polarizer (SCP) for which an International Patent is pending. The SCP is made by a sample of the same mixture of cholesteric liquid crystals. The concentration of each cholesteric component of the mixture can be adjusted in order to choose the spectral position of the rotatory dispersion curve. When used alone its effect has already been shown in the former section: it acts as rotator of the optical polarization with a strongly dispersive effect.

When used as a light modulator, it is placed between two polarizers which are adjusted in order to give minimum transmission if no stress is applied. Figure 4 shows the experimental set-up used to check the properties of this light modulator. An incoherent light source was used (lamp + monochromator) with a spectral bandwidth  $\Delta\lambda = 0.5 \text{ \AA}$ . The light transmitted was detected by a 56 CVP photomultiplier. A square-wave signal, ranging from negative to positive values, was

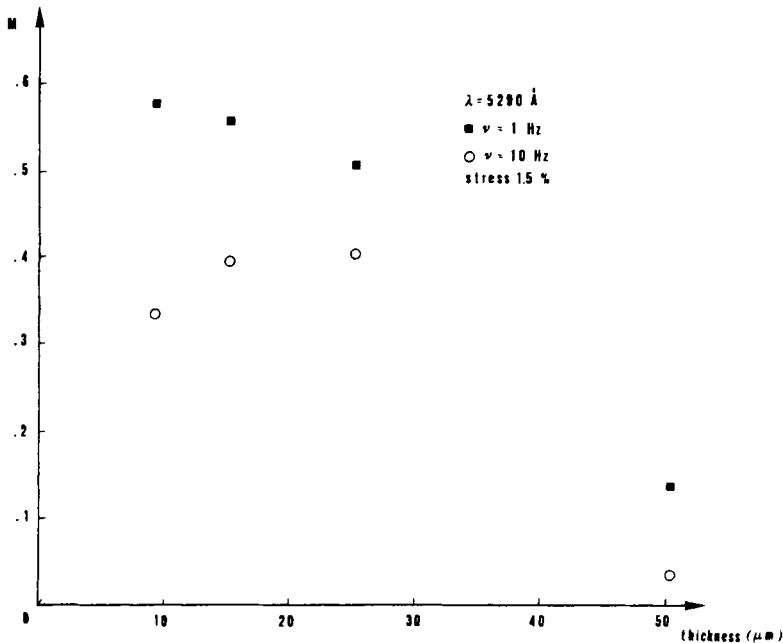


FIGURE 7 Light modulation  $M$  vs. sample thickness.

applied to the piezoelectric driver in order to get dilation and compression of the sample and obtain the maximum variation of the optical signal.

Measurements were performed on samples of different thickness looking for the optimum condition: maximum variation of the rotatory power and maximum extinction ratio between the two opposite polarizations.

As usual, we have defined the optical modulation as  $M = (I_{\max} - I_{\min})/I_{\max}$  where  $I_{\max}$  is the maximum value of the transmitted signal and  $I_{\min}$  the minimum one.

The measurements were performed for different frequencies of the exciting square wave signal and for different wavelengths. The mixture used in these experiments had the reflection peak at  $\lambda_0 = 5350 \text{ \AA}$ .

In Figures 5 and 6 the optical modulation  $M$  is reported vs. the modulation frequency  $\nu$  for samples  $9 \text{ }\mu\text{m}$  and  $25 \text{ }\mu\text{m}$  thick respec-

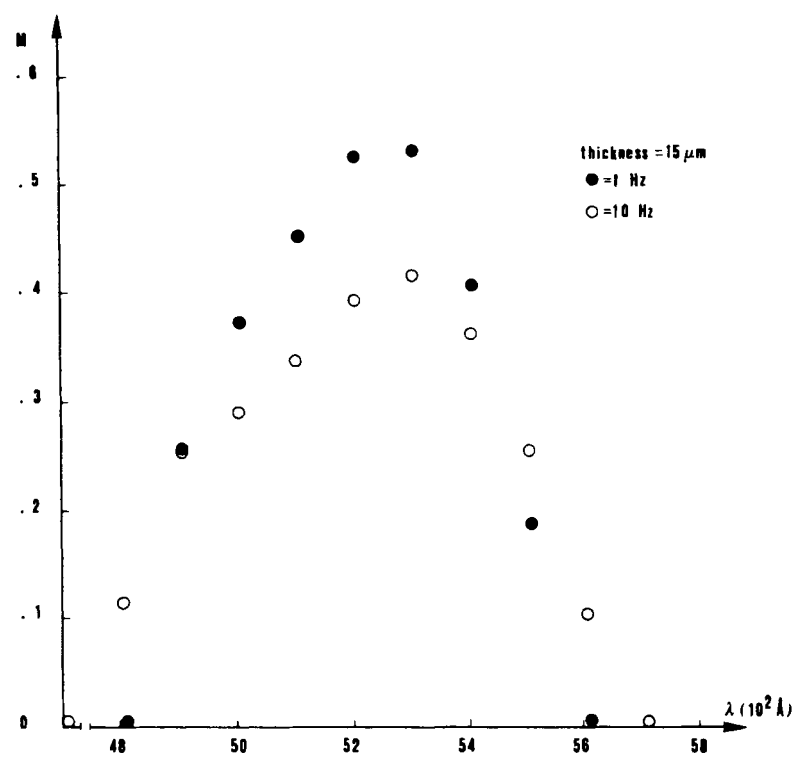


FIGURE 8 Light modulation  $M$  vs. the light wavelength.

tively. It is observed that using thin samples a slightly higher modulation is achievable. Increasing the exciting frequency we observe that  $M$  starts to decrease at the same  $\nu$  value for all the samples ( $\sim 6$  Hz) but the slope is steeper for thinner samples: we know that this is typical for a viscoelastic relation.<sup>11</sup> This behavior is well explained by Figure 7 where the modulation  $M$  is reported vs. the sample thickness for two frequencies, the first one lower and the second one higher than the cut-off frequency.

The wavelength dependence of the modulation  $M$  is reported in Figure 8 for a mixture  $15\text{ }\mu\text{m}$  thick. This figure confirms the peculiar dispersive properties of this device as already pointed out for the Voltage Controlled Polarizer (VCP).

### TUNING A DYE LASER BY SCP

As already published, the VCP has been recently used to get an interesting tuning effect in a dye laser cavity.<sup>12,13</sup> The same experiment has been carried out placing the new SCP device in the same

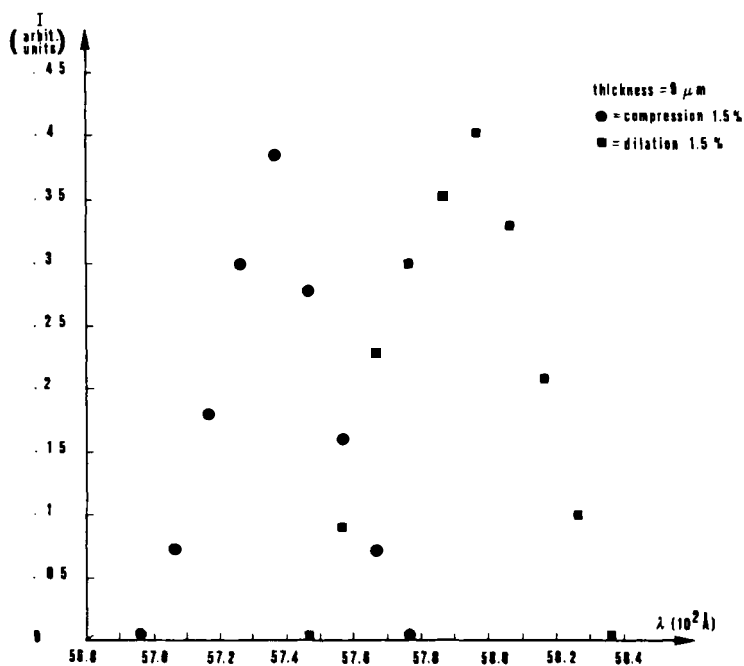


FIGURE 9 Dye laser lineshape for two different strain conditions of the S.C.P. in the cavity.

dye laser cavity. The principle of operation is the same as the VCP: the device (including two crossed polarizers) is used as a tunable transmission filter in order to obtain a modulation of the gain of the dye laser. Compression or dilation shifts the rotatory dispersion curve as illustrated in Figure 2. This effect changes the value of the wavelength which suffers a  $\pi/2$  rotation when transmitted by the liquid crystal, thus changing the transmission band of the whole device.

The advantage that can be expected by using the SCP instead of VCP is the possibility to control the tuning directions: i.e. we have a blue shift when the sample is compressed and a red shift when the sample is dilated. Moreover we should have a wider tuning range compressing and dilating the sample.

This is clearly demonstrated by our measurements. Typical laser linewidth spectra are reported in Figure 9. The dye laser used is a Rd-6G pumped by the second harmonic ( $\lambda = 5300 \text{ \AA}$ ) of a Nd-YAG Q-switched laser (pulse duration  $\tau_p = 20 \text{ ns}$ ) with the SCP placed in the same cavity which has been described elsewhere.<sup>12,13</sup> Care was

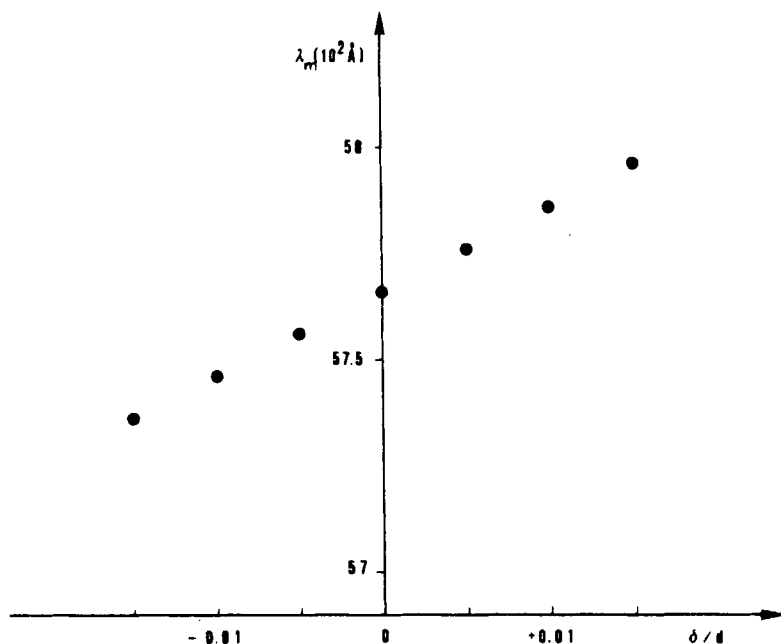


FIGURE 10 Peak wavelength of the dye laser line vs. the excitation  $\delta/d$  applied to the S.C.P.

taken in order to match the rotatory dispersion curve of the C.L.C. with the fluorescence band of the dye. The data reported in Figure 9 show features more interesting than the previously reported ones. In fact in this case the wider operation range allows us to have an almost complete separation of the laser lines for the extreme conditions of maximum dilation and maximum compression.

The tuning is completely reversible and linear with excitation. This is shown in Figure 10 where the wavelength  $\lambda_m$  corresponding to the maximum intensity of the laser line is reported vs. the applied strain.

We believe this device can find actual applications in laser tuning if some improvement will be done to obtain a narrower laser line-width.

The possibility to have a laser line sweep at about 10 Hz is an interesting feature and more detailed experiments on this application will be reported elsewhere.

## CONCLUSIONS

We have reported the effect of normal strains on the rotatory power of a Cholesteric Liquid Crystal. We have shown that De Vries' theory can be easily extended to take into account the mechanical effect if only elastic deformations are considered.

The experimental data are in good agreement with these previsions. Following this model, we have presented the modulation properties of a new optical device, that we have called Stress Controlled Polarizer (SCP), which promises more reliable applications than the Voltage Controlled Polarizer (VCP). This is due to the following characteristics of the SCP:

- a. fully linear behavior in the elastic regime
- b. easy control of the sign of the variations of the rotatory power
- c. ability to exploit a wide range of variations of the rotatory power passing from dilation to compression of the sample.

Preliminary results on the use of this device as a tuner when placed in a laser cavity are also reported and show a tuning range wider than that previously reported using a VCP.

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