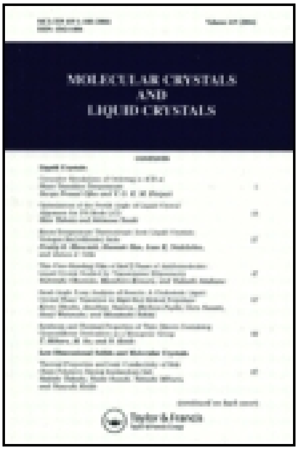

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Dynamical Response of Cholesteric Liquid Crystals to Mechanical Shearing Deformations

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We present experimental data on time-dependent behaviour of the apparent pitch of a Cholesteric Liquid Crystal (CLC) when it is submitted to mechanical shearing deformations. In particular we are interested to study the molecular reorientation process after the application of the shear. A good agreement is found between the calculated value of the characteristic time of this process, from a simple model, and the experimental one.

Keywords: shear, cholesteric L.C.

INTRODUCTION

The dynamical behaviours of layered liquid crystals (smectics and cholesterics) are very interesting to study because of their anisotropic properties, but it is very hard to obtain good experimental data, especially in cholesterics, because the physical situation is not always clear at all. We developed an experimental set-up[1,2] that by optical method (Bragg reflection) can be used to study the time-dependent behaviour of a planar CLC under external fields. We have already investigated the dynamical response of CLC under dilative stresses,[3,4] a d.c. electric field,[5] and mechanical shear deformations.[6]

In this work we present a quantitative report of our studies on the dynamical behaviour after the application of a step-like shearing deformation. In a previous paper[6] we showed that the apparent pitch is initially larger and finally less than the unperturbed one and also we pointed out that the mechanism determining these behaviours should be the competition between the unwinding of the helix structure and the tilt of the molecular layers.

Now we are interested to the molecular reorientation process.

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EXPERIMENTAL SET-UP

The experiment were performed on a planar structure of a cholesteric mixture whose unperturbed pitch was 4500 ± 25 Å at room temperature and it was proved to be both particularly pressure sensitive and temperature independent ($\Delta P/\Delta T <$ 5 Å/°C).

The sample, of 50 ± 5 μm thickness was sandwiched between two parallel optical glasses, of 1 cm of lateral dimensions, coated with a silane polymer (SURFINE) to fix the planar orientation, to the boundaries, in the x direction. The lower optical glass was fixed and the upper was moved by a loudspeaker, with low mechanical impedance, driven by a wave-form generator.

The parallelism of glasses was controlled, by optical method, to be better than 10^{-3} rad.

Our apparatus used to perform the experiments is schematically sketched in Figure 1: a while light beam, generated by a halogen lamp, falls onto the CLC sample with an external incidence angle of about 30°. The Bragg's reflection was collected and focused on the slit of a Jarrel-Ash monochromator. At the lateral window of the monochromator, the diffracted beam has a spectral width dependent on the input focus. In our case the width is 250 Å, about the same as the width of the Bragg reflection of the CLC sample. The spectra were detected by a photodiode array, then read by an electronic scanner and finally addressed to a buffer memory.

We were able, by a microcomputer, to record 24 spectra and to vary the time interval between two successive spectra from 9 msec to very long time in order to explore different time regions.

For further descriptions of experimental apparatus see References 1 and 2.

RESULTS AND DISCUSSION

In our experiments we have made measurements of the relative variations of apparent pitch with respect to time for various steplike shear deformations (in the range $S = 10 \div 500$ μm of shearing amplitude).

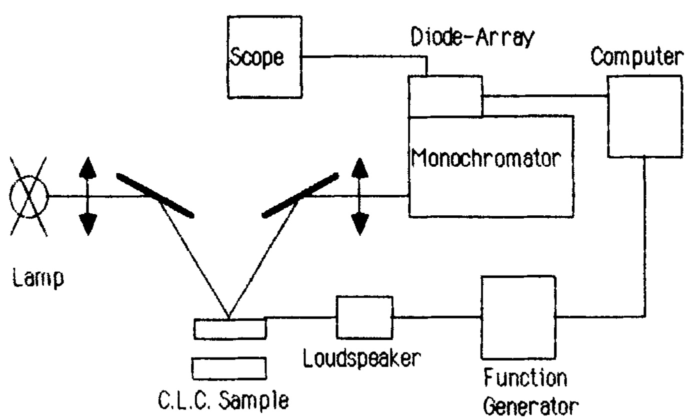

FIGURE 1 Schematic sketch of experimental apparatus.

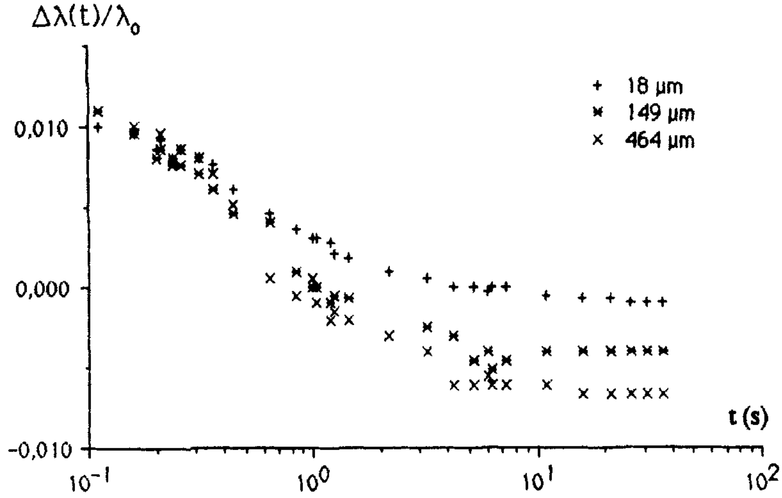

FIGURE 2 Behaviour of the relative variation of apparent pitch $\Delta\lambda(t)/\lambda_0$ vs. time for three different shearing amplitudes 18, 149 and 464 μm.

In Figure 2 we show the behaviour of the relative variation of apparent pitch $\Delta\lambda(t)/\lambda_0 = (\lambda(t) - \lambda_0)/\lambda_0$ vs. time (where λ_0 is the wavelength of the maximum reflection at rest and $\lambda(t)$ is the wavelength of the maximum of reflection measured, after application of the steplike shear deformation, at different times) for three different shearing amplitudes (18, 149 and 464 μm).

This behaviour has been already qualitatively explained in Reference 6.

Now, our interest is restricted to study the reorientational relaxation process, i.e., the process of molecular reorientation that occur in the range of time between 10^{-1} and 10^{1} sec.

In Figure 3a we evidence this temporal region for the same three different shearing amplitudes of Figure 2.

The experimental data, in the region of interest, can be interpolated by weighted χ^2 method (because different temporal density of experimental data), by an exponential law that give us the experimental characteristic time of the process (see Figure 3b).

In Figure 4 are presented the experimental characteristic times for the different shearing amplitudes used in our experiment.

From the Figure 4 is possible to observe that these characteristic times are, within the errors, constant, i.e., are independent from shearing amplitudes but depend only on the material. From our data this characteristic constant time comes out about 0.6 ± 0.1 sec.

As we have previously written[1,6,7] besides the unwinding process there is the tilt of molecular layers that contributes to the pitch variation in opposite sense.

In previous papers[1,7] we have shown how that contribution is negative and such a variation is relatively smaller than that due to unwinding.

In fact, in the latter case we have an effective increasing of the pitch, instead in the former one the shortening is only apparent.

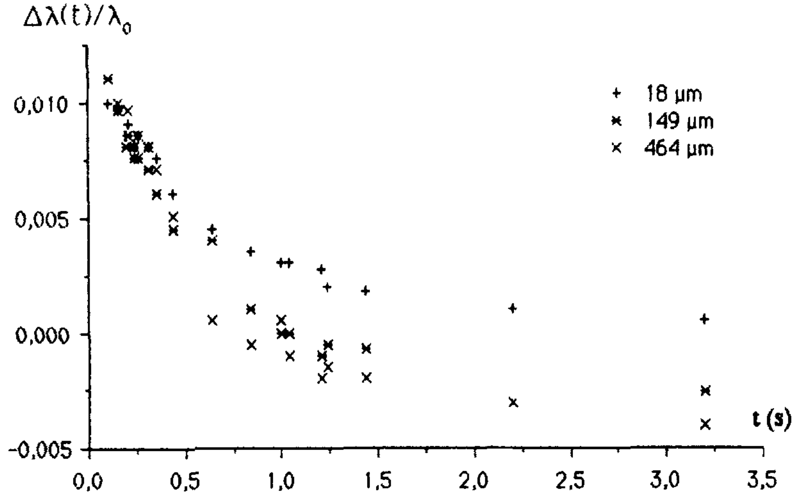

FIGURE 3a Behaviour of the relative variation of apparent pitch $\Delta\lambda(t)/\lambda_0$ vs. time in the temporal region where occurs the process of molecular reorientation. The shearing amplitudes are the same of Figure 2. The experimental data, for every shearing amplitudes, can be interpolated by an exponential law.

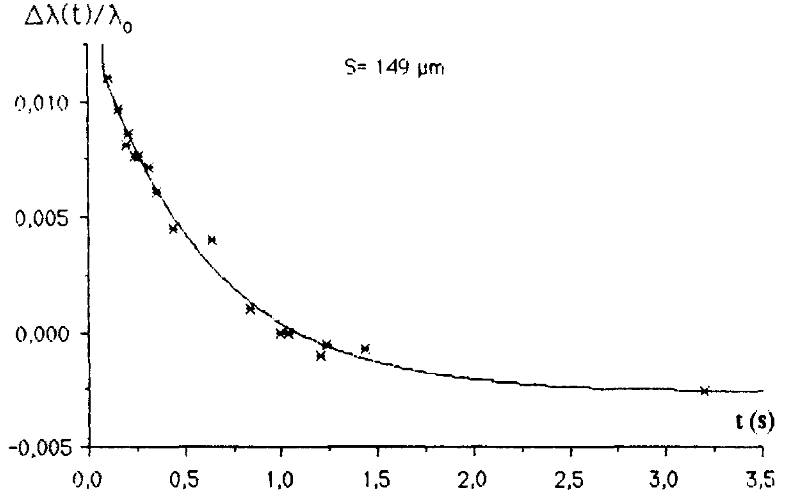

FIGURE 3 Bis Experimental data for shearing amplitude S = 149 μm interpolated by weighted χ^2 method by an exponential law that give us the experimental characteristic time of the process.

Let us discuss the two contributions in a separate manner.

The dynamical equation describing the unwinding process is[8]:

$$K_2 \frac{\partial^2 \phi(z,t)}{\partial z^2} = \gamma_1 \frac{\partial \phi(z,t)}{\partial t} \tag{1}$$

where $\phi(z,t)$ is the azimuth of the director, z is the axis parallel to the helical axis of unperturbed CLC.

Then, as an order of magnitude estimate, the relative relaxation time is:

$$\tau_u \approx \frac{\gamma_1}{K_2} \frac{1}{q_u^2}$$

where $q_u \approx (2\pi/\ell)$ is the fundamental wavevector and ℓ is a characteristic length of the same order of layer dimension.

Using the following characteristic values[7,8,9] $K_2 \approx 5\ 10^{-7}$, $\gamma_1 \approx 0.9$ in c.g.s. and taking $\ell \approx 10^{-4}$ in c.g.s., we obtain a value of $\tau_u \approx 10^{-3} \div 10^{-2}$ sec.

The smallness of such a time indicates that the unwinding process does not play any role in the long time reorientational relaxation process.

As regard the tilt of the molecular layers, the equilibrium between elastic and viscous torques can be written as[10]:

$$K \frac{\partial^2 \psi}{\partial z^2} = \gamma_1 \frac{\partial \psi}{\partial t} + \frac{1}{2}(\gamma_1 + \gamma_2) \frac{\partial v_x}{\partial z} \tag{2a}$$

where K is an average value between K_1 and K_3 and ψ is the angle between the helical axis of CLC and the z-axis (see Figure 5).

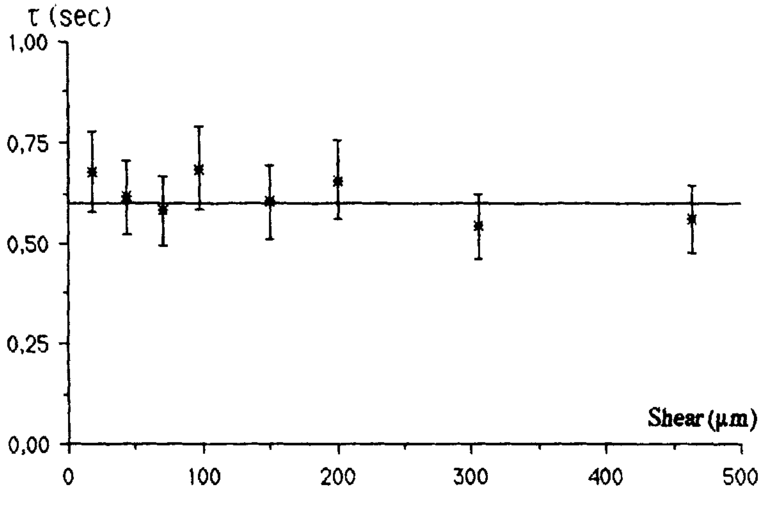

FIGURE 4 Experimental characteristic times vs. shearing amplitudes. The typical relative error is about 15%. The average value, denoted by full line, is about 0.6 sec.

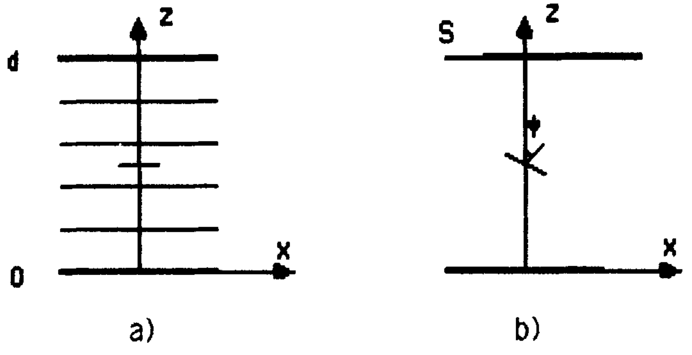

FIGURE 5 Configurations of the model of the CLC: a) unperturbed cholesteric with the helical axis parallel to the z axis and layers in the plane x–y. b) Sheared cholesteric where ψ is the angle between the helical axis and the z axis, S is the shearing amplitude. The angle ψ is emphasized to make clear the tilt of helical axis.

Neglecting the inertial effects, the gradient $\partial v_x/\partial z$ is related to ψ by the Leslie equation of hydrodynamic motion[11]:

$$\frac{\partial}{\partial z}\left(\eta_1 \frac{\partial v_x}{\partial z} + \alpha_3 \frac{\partial \psi}{\partial t}\right) = 0 \tag{2b}$$

where η_1 represent the equivalent kinematic viscosity for a CLC shear flow.

In order to solve the system of equations (2) the following boundary conditions should be considered:

$$\begin{aligned} \psi(z = 0) &= \psi(z = d) = 0 \\ v_x(z = 0) &= v_x(z = d) = 0 \end{aligned} \tag{3}$$

Looking for a solution of the form:

$$\psi(z,t) = \hat{\psi}(z)e^{-\omega t}$$

the solution of the system of equations (2), together with the boundary conditions (3), reduces to a Sturm-Liouville problem.

The corresponding eigenvalue is:

$$|\omega| = \left(\frac{2\pi}{d}\right)^2 \frac{K\eta_1}{\gamma_1\,\eta_1 - (\gamma_1 + \gamma_2)\,\alpha_3} \tag{4}$$

so that, using the following value[7,8,9] $K \approx 10^{-6}$, $\eta_1 \approx 5\ 10^{-1}$, $\alpha_3 \approx -10^{-2}$ and $\gamma_2 \approx -1$ we obtain:

$$\tau_t = |\omega|^{-1} \approx 10^0 \text{ sec.}$$

in agreement with the experimental characteristic time of reorientation relaxation process.

CONCLUSIONS

The aim of this work was to investigate the experimental molecular reorientation process following the application of a step-like shear along an axis perpendicular to helical axis of a CLC in planar configuration.

We showed that the characteristic time of this phenomena is independent from shearing amplitudes and depends only on material properties.

We have tried to understand the probable mechanism which gives rise to the relaxation of CLC. In particular, we have identified two phenomena: the unwinding of the helix structure and the tilt of the molecular layers.

Assuming for simplicity that the two phenomena are independent between each other, we have tried to model them, in a rather crude manner.

Due to the fact that the characteristic lengths involved are rather different for the two phenomena, we found that the time requested for the tilt of molecular layers is much longer than that for unwinding of the helix structure.

Finally, we have attributed to the tilt of molecular layers the observed relaxation process.

Much more experimental measurements are in order, particularly directed towards the study of the relaxation process against the thickness of the sample. A future paper will be devoted to such work, together with a better theoretical treatment of the phenomena.

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