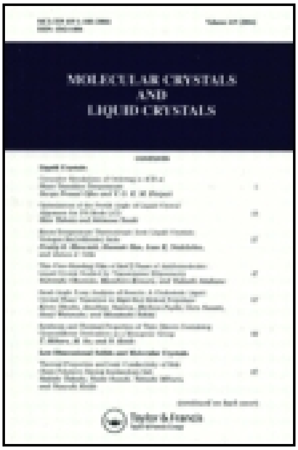

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Oscillations in Layered Liquid Crystals

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Oscillations in Layered Liquid Crystals

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We present some experiments on cholesteric liquid crystals mechanically stressed perpendicularly to the layers by a step-like dilation. Results show the existence of a decrease of the apparent pitch on very short time scale with respect to permeative and hydrodynamic times. A conjecture on such phenomenon is also presented.

Keywords: cholesteric liquid crystals, hydrodynamics, oscillations

INTRODUCTION

Smectic (SLC) and cholesteric (CLC) liquid crystals (layered liquid crystals LLC) exhibit interesting dynamical and mechanical properties.[1]

More precisely, they can show solid-like elastic behavior and they can sustain strains applied perpendicular to the layers. Indeed, a pressure gradient across the layers should relax by permeative flow toward a new equilibrium state.[2–6] Furthermore it is well established that in the high frequency regime, elastic waves can be excited in LLC leading to the well known effects of first and second sound.[1] Such phenomena happen on quite different time scales: $\tau_c \ll \tau_p \approx \tau_h$[3,4,6] ($\tau_c$, τ_p, τ_h are respectively the characteristic times associated with the elastic, permeative and hydroelastic regimes).

On the other hand, some experiments on CLC mechanically stressed perpendicularly to the layers by step-like dilation, seem to show the existence of a decrease of the apparent pitch on a time scale intermediate between τ_c and τ_p; the latter regime appears to be more visible in thinner samples. Such effects are not in agreement with the experiments previously made in the static case.[7]

In this paper we first give a detailed analysis of our experiments, then we briefly conjecture on a possible explanation for the observed phenomenon.

EXPERIMENTAL SET-UP

The experiment was performed on a planar structure of a cholesteric mixture (31.2 wt% of cholesteryl chloride, 21.1 wt% of cholesteryl nonanoate and 47.7 wt% of cholesteryl oleyl carbonate) which is particularly pressure sensitive and temperature independent in a large range of temperatures. The unperturbed pitch is $P_o = 4600 \pm 25$ Å at room temperature.

The samples were sandwiched between two parallel glass plates coated with a silane polymer (Surfine) to fix the planar orientation. The parallelism of the glass plates was checked by optical methods to be better than 10^{-3} rad. The thickness d of the sample was initially determined by Mylar spacers removed in the course of the experiment. Both plate parallelism and sample thickness were also monitored by optical method after removal of the Mylar spacers. To produce the dilation, we seal one of the glasses to a piezoelectric element driven by a step-like voltage at a frequency of 10^{-3} Hz.

In a coarse-grained picture, our sample can be regarded as a layered system with a layer spacing $P_o/2$, where P_o is the unperturbed helix pitch and $L = 4$ cm is the sample width (see Figure 1), so we can use optical techniques to study deformations of the CLC structure.

The experimental setup is schematically described by Figure 2: a white light beam impinged on the sample at an angle of about 30°. The Bragg reflection beam was collected and analysed by a Jarrel-Ash Spectrometer (with a resolution of ± 3 Å).

The spectra were detected by a photodiode array placed behind the lateral window of the monochromator (the detectable spectral width was 250 Å, about the same as the width of the Bragg reflection of the CLC sample), then read by an electronic scanner and finally addressed to a buffer memory which could store up to 24 different spectra. The synchronization between sample excitation and data acquisition was provided by a Commodore 128 microcomputer. For further description of the experimental apparatus see Reference 8.

The time evolution of the relative variation of apparent pitch for five different thicknesses d of the sample are shown in Figure 3 a–e.

For every thickness d we fixed the relative deformation $\epsilon = (\delta/d) = 10^{-2}$ (δ is the thickness variation induced by piezoelectric ceramic), i.e. it remains in the region of linear deformation of CLC as we have already shown.[9]

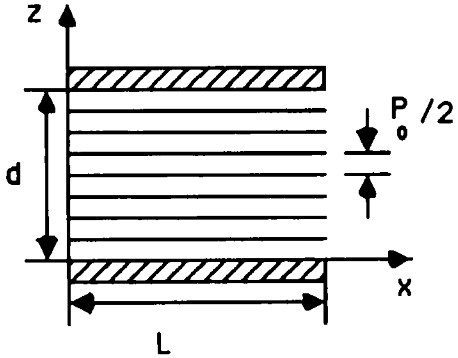


FIGURE 1 Schematic drawing of the geometry of the experiment. The sample of thickness d is homogeneously aligned along the x-axis at the boundaries. $L = 4$ cm is the sample width.

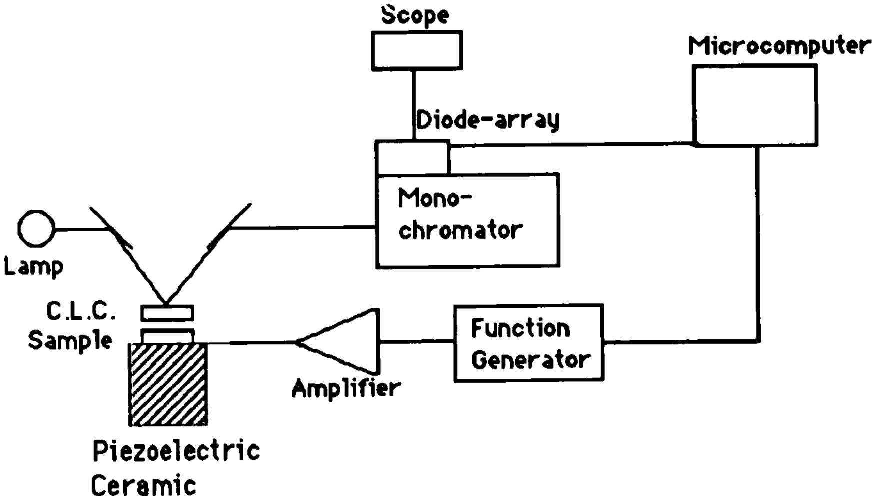
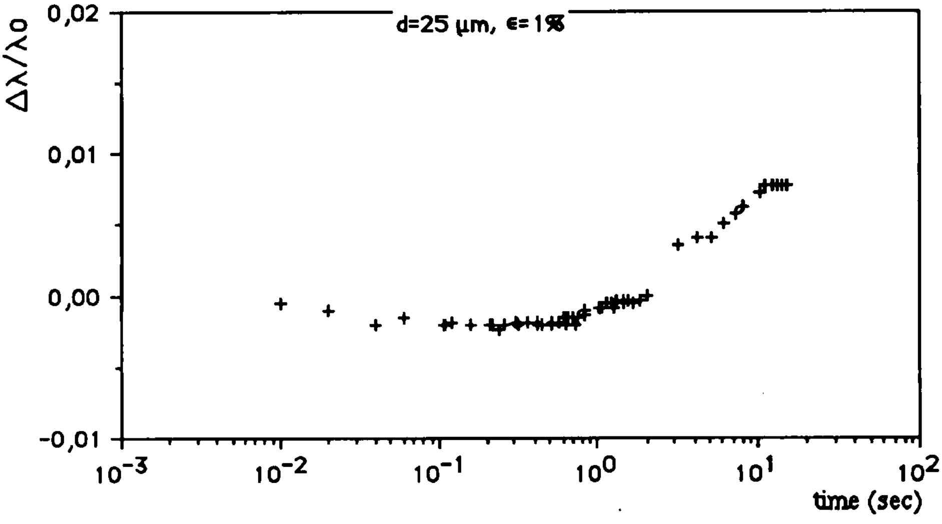


FIGURE 2 Schematic sketch of experimental apparatus.

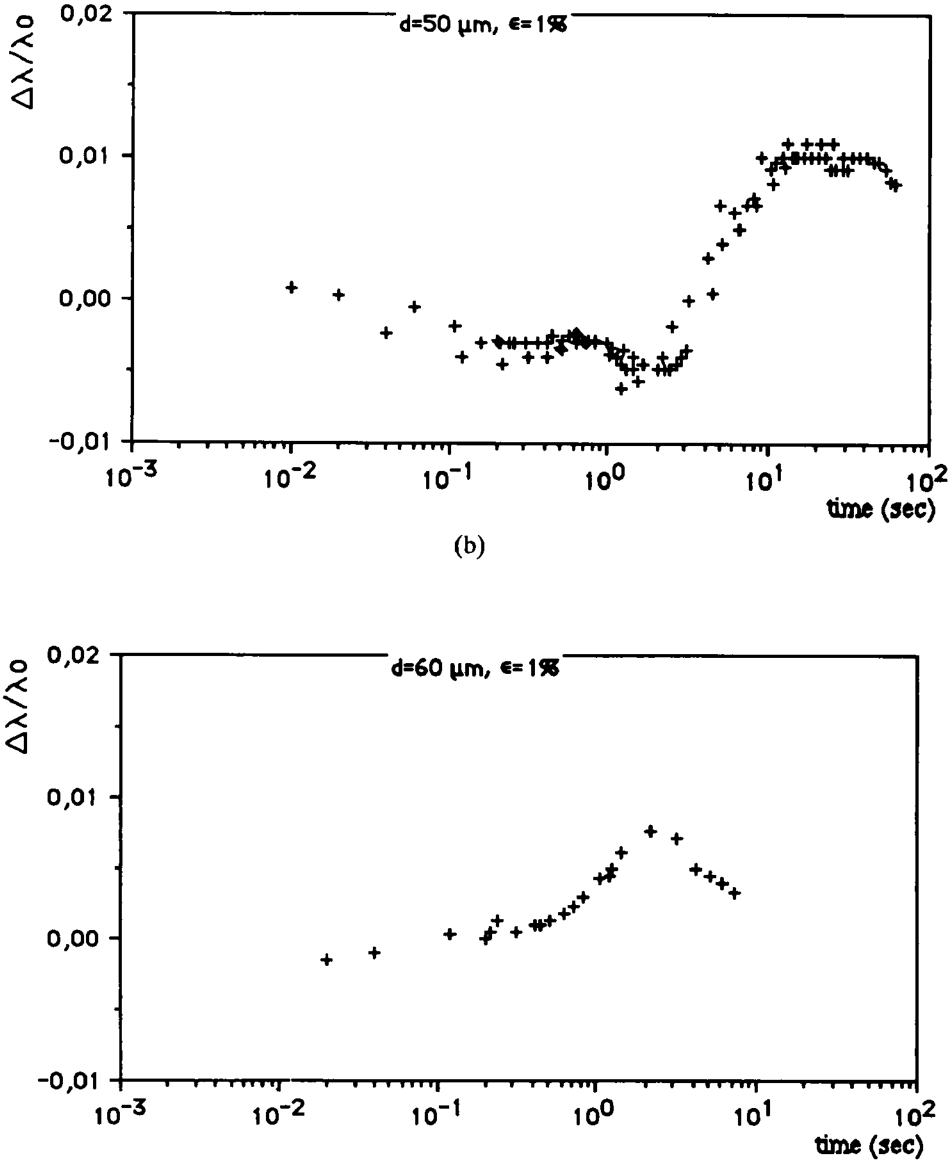
Using the experimental set-up, we can easily distinguish two main phenomena described earlier: the decay due to permeation at times of the order of 1–10 s, and the new equilibrium reached at times of the order of 10 s or more.

On the other hand, mainly for thin samples, an oscillation of apparent pitch, is easily visible at times of the order of 10^{-2} sec. This effect is the first response of



(a)

FIGURE 3 (a–e) Experimental behavior of the relative variation of apparent pitch $\Delta\lambda/\lambda_0$ vs. time for sample thicknesses 25, 50, 60, 100 and 175 µm respectively. For every thickness the relative deformation is $\epsilon = (\delta/d) = 10^{-2}$.

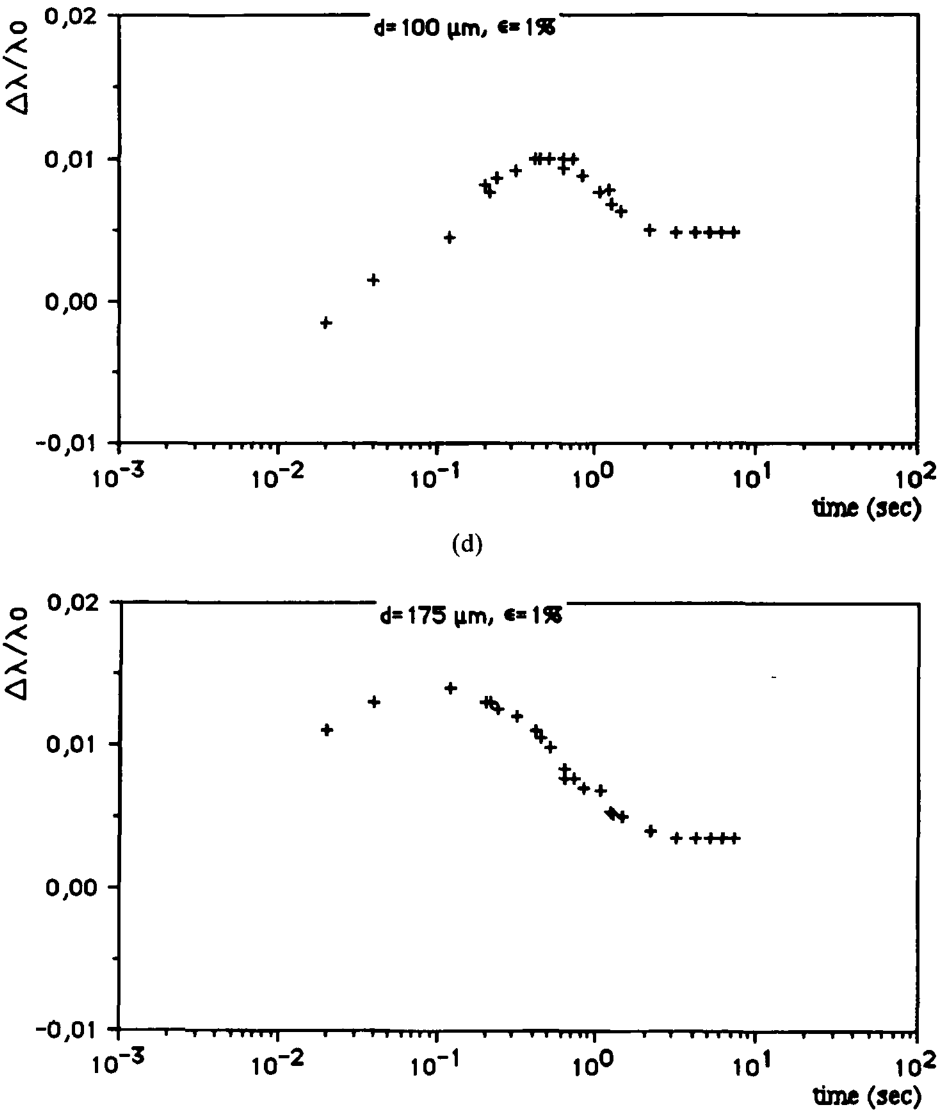


(b)

(c)

FIGURE 3 (*continued*)

the sample to the step-like dilation observed in our experimentally accessible times. We must consider that the step-like dilation rises in a time of the order of 10^{-3} s or less, which is smaller than the shortest observable time by our experimental techniques.



(d)

(e)

FIGURE 3 *(continued)*

Moreover, the thinner the sample, the more visible oscillation effect will result. Finally, for thin samples, the decay due to permeative effects is considerably reduced with respect to large samples.

DISCUSSION AND CONCLUSION

Permeative effects in LLC, due to coupling between the flow in the Z-direction and layer distortion are responsible for the response time of the variation of the apparent pitch. This phenomenon depends on a predetermined value of the viscosity.[6]

The characteristic permeative and viscous decay times[2] can be estimated to be of the order of:

$$(\tau_p)^{-1} = \lambda_p \cdot B \cdot \left(\frac{2\pi}{d}\right) ;$$

$$(\tau_h)^{-1} = (2\pi)^2 \cdot \nu \cdot \left(\frac{1}{d^2 + L^2}\right)$$

where L is the width sample, λ_p is the permeative constant, B is the equivalent layer compression modulus and ν is the kinematic viscosity. Using the following numerical values[1]: $B = 10^4$, $\lambda_p \cdot B = 1.5\ 10^{-6}$ and $\nu = 0.65$ (in c.g.s.), we found that, for the thicknesses range we have studied (i.e. $25\ \mu\text{m} \leq d \leq 175\ \mu\text{m}$), the characteristic times τ_p and τ_h are of the order of a few seconds.

We want to investigate oscillations on frequencies up to 10^2 Hz., so that we can disregard the permeative and viscous effects.

In the incompressible case, disregarding nonlinear effects and assuming symmetry around the Z-axis (see Figure 1), the hydrodynamic equations describing an LLC sample reads:

$$\frac{\partial V_z}{\partial Z} + \frac{\partial V_x}{\partial X} = 0 \tag{1}$$

$$\rho \frac{\partial V_x}{\partial t} = - \frac{\partial p}{\partial X} \tag{2}$$

$$\rho \frac{\partial V_z}{\partial t} = - \frac{\partial p}{\partial Z} + B \frac{\partial^2 u}{\partial z^2} \tag{3}$$

where $(\partial u/\partial t) = V_z$ (u being the layer's displacement), $\rho(\approx 1)$ the density and V_X, V_z are the flow velocities in the X and Z direction respectively.

Eliminating the pressure terms, Equations 1–3 reduce to:

$$\frac{\partial^2}{\partial t^2}\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial z^2}\right) = c^2 \frac{\partial^2}{\partial z^2}\frac{\partial^2 u}{\partial x^2} \tag{4}$$

where $c = (B/\rho)^{1/2}$ is a characteristic velocity of propagation of the perturbations. Equation 4 describes the propagation of elastic waves.

Suppose, first of all, that the variations in the X and Z directions was of the same order of the sample thickness d:

$$\left|\frac{\partial^2 u}{\partial x^2}\right| \approx \left|\frac{\partial^2 u}{\partial z^2}\right| \approx \frac{u}{d^2}$$

In such a case, using a Fourier analysis of the layer's displacement

$$u(x,z,t) = \sum_{q_\perp q_\parallel} \tilde{u}(q_\perp, q_\parallel, \omega)\, e^{+i(q_\parallel z + q_\perp x - \Omega t)}$$

we obtain oscillations of u with a frequency Ω given by

$$\Omega^2 = c^2 \frac{q_\perp^2 q_\parallel^2}{q^2} \quad (q^2 = q_\perp^2 + q_\parallel^2)$$

and a velocity $C_2 = (\Omega/q)$. The time scale is of the order of $\tau \approx (d/c)$, which, in the thickness range studied, is of the order of 10^{-4} s. These oscillations are usually referred to as second sound.[1] Since in our case $L >> d$, it is obviously not possible to observe second sound.

A quite different situation is obtained when variations in the X and Z directions are different, as is the case here. Indeed, suppose that

$$\left|\frac{\partial^2 u}{\partial x^2}\right| << \left|\frac{\partial^2 u}{\partial z^2}\right|$$

so that $(d/L) << 1$ and the characteristic times are of the order of $\tau \approx (L/c)$.

In such a case, the second term on the left hand side of Equation 4 balances the right hand side and Equation 4 becomes:

$$\frac{\partial^2 u}{\partial t^2} = c^2 \frac{\partial^2 u}{\partial x^2} + f(x,t)\, z + g(x,t) \tag{5}$$

This equation has the form of a nonhomogeneous cylindrical oscillatory equation, whose solution can be written as:

$$u(x,z,t) = \frac{1}{2c} \int_0^t d\tau \int_{x - c(t - \tau)}^{x + c(t - \tau)} d\xi\, F(\xi,z,\tau)$$

where $F(\xi,z,\tau) = f(\xi,t) \cdot z + g(\xi,t)$.

For simplicity, we look for solutions of Equation 5 which behave according to:

$$F(\xi,z,t) = (A\, z + K) e^{i(q_\perp \xi - \omega t)}$$

where A and K are arbitrary constants. After some straightforward calculation we find:

$$u(x,z,t) = \frac{(Az + K)}{2\,\omega_\perp\,(\omega_\perp - \omega)}\, e^{i(q_\perp x - \omega t)}\left[1 - e^{-i(\omega_\perp - \omega)t}\right]$$

$$X\left\{1 + e^{2i\omega t}\left(\frac{\omega_\perp - \omega}{\omega_\perp + \omega}\right)\left[\frac{1 - e^{i(\omega_\perp - \omega)t}}{1 - e^{-i(\omega_\perp - \omega)t}}\right]\right\} \qquad (6)$$

where $\omega_\perp = q_\perp \cdot c \approx 10^2$ Hz is a characteristic frequency (for the wavevector we reasonably take $q_\perp \approx 2\pi/L$).[6]

The main contribution to the time variation of the layer displacement in Equation 6 comes from the resonance frequency $\omega \approx \omega_\perp$. Introducing a small parameter ψ such that

$$|\omega - \omega_\perp| \approx \psi << 1$$

we find at the zero order:

$$u = \frac{(Az + K)t}{2\,\omega_\perp}\, e^{i\,[q_\perp(x - ct) - 1]} \qquad (7)$$

It is worthwhile to note that the possibility of appearance of the oscillations is more probable for thin samples with respect to large samples. Indeed when the condition $(d/L) << 1$ is better satisfied, the Equation 5 is more realistically the "true" equation which describes the oscillatory phenomenon.

Really in our experiments we observe an oscillation at times of the order of $10^{-2} \leq \tau \leq 10^{-1}$ s, and such oscillation is more visible when the condition $(d/L) << 1$ is better satisfied. Then we tentatively conjecture that the "strange" behavior observed for thin samples was due to the above-described phenomenon: namely oscillation response of the structure, to external forces, at times of the order $\tau \approx (\omega_\perp)^{-1}$.

Finally some considerations are in order. Really it is difficult to satisfactorily bind the experimental evidences to the conjecture we have made, because some physics has been disregarded and some simplifications are taken into account. Yet some other physical effects could be improved to explain the behavior seen in the experiments.

Nevertheless the possibility of seen the oscillations remains an open question.

Further theoretical considerations and experimental measurements are under way to better clarify this phenomenon.

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