Permeative Flow in Cholesteric Liquid Crystals

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A planar texture of a cholesteric liquid crystal is submitted to a uniaxial steplike strain parallel to the helical axis. Using a time-resolved optical Bragg-scattering technique, we measure the average pitch of the texture (and its dispersion) versus time. We observe a nonuniform hydroelastic dilation, which relaxes to the uniform-strain equilibrium state through a permeative flow.

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Layered liquid crystals (smectics and cholesterics) present very anisotropic mechanical and dynamical properties[1]: For flow parallel to the layers, they behave as 2D fluids; on the other hand, they can sustain normal strains perpendicular to the layers. A pressure gradient across the layers should relax by a weak "permeative" flow, as suggested by Helfrich.[2] It is difficult to observe a macroscopic permeative flow, if only because of the existence of additional plastic flow associated with defect motion.[3] An interesting method to observe permeation would be to create a nonuniform strain in an homogeneous sample, and to observe its relaxation to the uniform equilibrium texture. In smectic liquid crystals, these time-dependent strains are *a priori* difficult to observe, because the layer periodicity is in the x-ray range. Cholesteric liquid crystals are much better suited for such an experiment since their spatial period, the pitch of the helical texture, falls in the optical range. In this Letter we report on the first observation of the time dependence of the pitch of a cholesteric liquid crystal submitted to an instantaneous steplike strain normal to the layers.

In a previous model, the frequency and space dependence of the normal distortion of smectic layers under a periodic strain was calculated.[4] We can adapt these results to cholesterics since we know[5] that when the pitch p of the helix is much smaller than $2d$, the sample thickness, cholesterics behave like smectics, with an equivalent layer compression modulus $B = Kq_0^2$, where K is the twist curvature constant of the cholesteric and $q_0 = 2\pi/p$. Let us consider an ideal cholesteric planar texture [Fig. 1(a)] between two plates separated by $2d < L$ (L is the sample width). The plates can be moved quasi instantaneously by amounts $\pm\delta$, parallel to the helical axis. This results in an external dilative strain $\epsilon = \delta/d$. After a long relaxation time, the equilibrium state of the texture should be a uniform dilation with a layer normal displacement $u = z\delta/d$ and $\partial u/\partial z = \epsilon$ [the $\vec{z}$ axis is normal to the layers, see Fig. 1(a)].

For shorter times, it was shown in Ref. 4 that the layers are frozen inside the material. The layer profile is then imposed by the hydrodynamical flow V (V_x, V_z). This flow is locally of a Poiseuille kind, $V_x \sim (z^2/d^2 - 1) f(x)$. Because of the incompressibility, $\partial V_z/\partial z \sim 1 - z^2/d^2$. As the layers are frozen in the material the layer velocity is $\partial u/\partial t = V_z$, and the z profile of u is given by $u = \frac{3}{2}\delta(z/d - z^3/3d^3)$ [Fig. 1(b)] because of the boundary conditions $u = \pm\delta$ for $z = \pm d$. The local strain $\partial u/\partial z$ is maximum in the center of the sample: $\partial u/\partial z = \frac{3}{2}\delta/d = \frac{3}{2}\epsilon$. This "hydroelastic" regime is the steady-state regime for frequencies $\omega_H \sim q_x^2 d^2 \times Kq_0^2/\eta$, where $q_x = \pi/L$ and η is a viscosity. ω_H is the damping frequency of the layer motion, in the absence of permeation, under the action of layer-curvature elasticity and viscous forces. Because $L \gg d$, ω_H is much smaller than the damping frequency Kq_0^2/η of the helix.

This hydroelastic regime should be observed at a delay time $\tau_H = 2\pi/\omega_H$ after the steplike strain. For low frequencies a permeative flow should also develop, in order to relax the nonuniform distortion toward the uniform distortion. The permeation equation is[1]

$$\dot{u} = \lambda_p B\, \partial^2 u/\partial z^2 = (K/\eta) q_z^2 u,$$

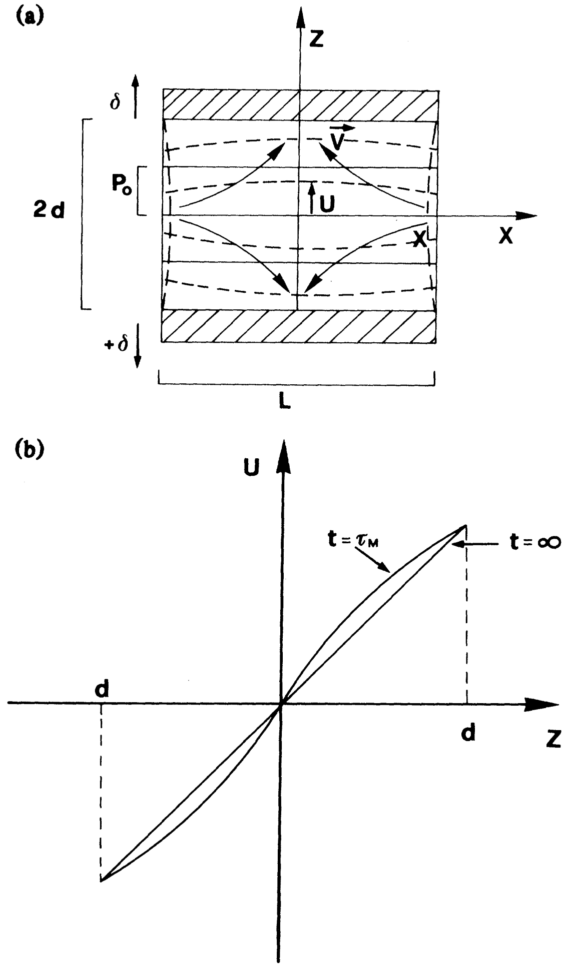

FIG. 1. (a) Experimental geometry: $2d$, sample thickness; L, sample width; p_0, unperturbed cholestric pitch; $\pm\delta$, imposed dilation (along z). The unperturbed sample is indicated in full lines; the nonuniformly perturbed (dilation) sample, in dashed lines. u is the layer distortion. The flow associated with dilation is indicated by arrows. (b) Thickness profile of a distortion u, in the center of the glass plate, for uniform relaxed texture ($t=\infty$) and a texture of cholesteric planes frozen in the shear flow associated with dilation ($t=t_M$).

since λ_p, estimated by Helfrich,[2] is written as $\lambda_p^{-1}=\eta q_0^2$. There should exist a permeation boundary layer[6] of thickness l defined by $(\pi^2/4l^2)q_xq_0$, i.e., $l^2=pL/8$.

For thin samples ($d<l$), $q_z=\pi/2d$ and the permeation damping rate is $\tau_p^{-1}=K\pi^2/4d^2\eta>\omega_H$. For thick samples ($d>l$), permeation is restricted to the boundary layer, with $q_z^2=q_xq_0=2\pi^2/pL$, and the permeation damping rate should be $\tau_p^{-1}=K2\pi^2/pL\eta<\omega_H$. For delay times shorter than τ_H, inertia is no longer negligible and "second sound" modes[1] could be excited.

The cholesteric sample we use is a room-temperature mixture of cholesterol nonanoate, chloride, and oleyl carbonate, with an equilibrium pitch $p_0=0.4$ μm, in a planar configuration. The glass surfaces are coated with a silane polymer (MAP) to fix the planar orientation. The sample thickness is chosen as $2d=75$ μm, with $L=4$ cm. This corresponds to $2l=90$ m, comparable with $2d$; i.e., the permeation boundary layers can extend across the entire thickness of the sample. Thicker samples are more difficult to obtain in well-aligned monodomains. One of the glasses is pulled by a piezoelectric ceramic an amount δ ($\delta<1$ μm), in steps of 100 sec. The sample is illuminated with a white-parallel-light source at an angle of incidence of 20°–30°. The Bragg-reflected light is analyzed with a monochromator followed by a BM Spektronix 512-diode-array system (AK 500), to perform time-resolved optical spectroscopy.[7] For each 100 msec delay, we obtain a spectrum of the Bragg-reflected light, characterized by its maximum-intensity wavelength p and width W.

In the absence of an applied dilation, p is found to be $p_0=0.4$ μm. $W_0=150$ Å corresponds to the instrumental resolution. In the presence of a sudden dilation δ, p depends on the time delay t after the dilation. We have plotted in Fig. 2 our $p(t)$ data, for two dilative strains $\epsilon=5\times10^{-3}$ and $\epsilon=10^{-2}$. The maximum shift $\Delta p(t)=p(t)-p_0$ is reached at $t_M\sim1$ sec, and then $\Delta p(t)$ decreases with a time constant $\tau\sim1.6$ sec toward a steady value $\Delta p(\infty)$, constant over more than 60 sec. For times shorter than t_M, our present data show a transient regime, with an apparant inversion of the strain. This regime, which could correspond to an inertial mode, must be analyzed in more detail and will be reported later. The associated spectral bandwidth $W(t)$ is also found to be maximum around t_M [$W(t_M)\sim170$ Å for $\epsilon=10^{-2}$ and relaxes back to $W_0=150$ Å with the same τ].

Varying ϵ, we have plotted (Fig. 3) the strain dependence of the relative pitch variation $\alpha=\Delta p/p_0$, for the maximum value at $t=t_M$ (α_M) and the steady value at large t (α_∞). We observe a linear dependence of α_M and α_∞ on ϵ. A least-squares fit gives $\alpha_\infty=(1\pm0.1)\epsilon$ and $\alpha_M=(1.4\pm0.1)\epsilon$. We can obviously interpret $\alpha=\Delta p/p_0$ as an averaged measurement of the local strain $\langle\partial u/\partial z\rangle$ of the cholesteric layers. $\alpha_\infty=\epsilon$ associated with $W=W_0$ is just the expected shift from a uniformly strained sample as previously observed.[8] $\alpha_M\sim1.4$ and the large $W(t_M)>W_0$ are signatures of nonuniformly strained cholesteric, with layers frozen in the flow, as previouly explained. In fact,

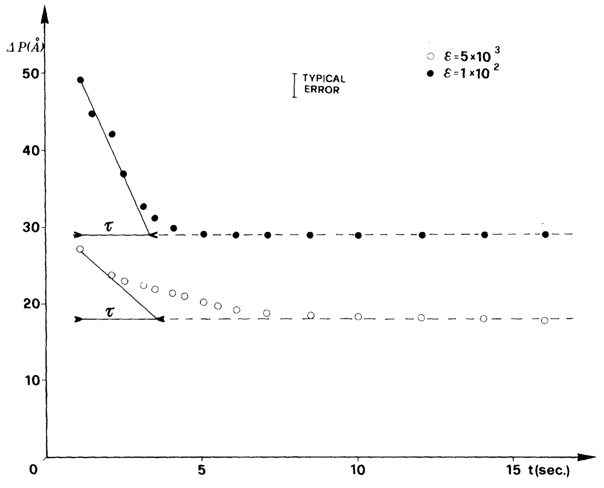

FIG. 2. Time evolution of the apparent pitch variation Δp for two different strains $\epsilon=\delta/d$, $\epsilon_1=5\times10^{-3}$ and $\epsilon_2=10^{-2}$.

the factor 1.4 instead of 1.5 probably results from the average effect of the more or less strained parts of the sample. To identify the mechanisms involved in the $p(t)$ evolution, we can compare the times observed in this experiment (t_M and τ) with the estimated τ_H and τ_p. As our sample is in the intermediate-thickness range ($d\sim l$), the hydroelastic and permeative times τ_H and τ_p should be equal. We can estimate τ_p; for instance, with $K\sim5\times10^{-7}$ cgs and $\eta\sim0.1$ cgs, we get $\tau_p\sim1$ sec. We then identify t_M with τ_H and the observed decay time τ with the permeation damping time, which controls the relaxation of the nonuniformly strained hydroelastic distortion towards the uniform purely elastic state of equilibrium.

In conclusion, we have analyzed the time evolution of the wavelength of maximum white-light Bragg-scattered intensity from a planar sample of a cholesteric liquid crystal submitted to a instantaneous steplike strain. Associating the relative Bragg-scattered wavelength shift with the local strain of the cholesteric layers, we have demonstrated the existence of two deformation states of the cholesteric texture: a long-delay-time uniform strain of the helix, matching the external dilation, and a shorter-delay-time nonuniform strain. This later strain, 50% larger than the applied external strain, can correspond to the predicted high-frequency "hydroelastic" regime, where the cholesteric layers are frozen in the macroscopic shear flow associated with the dilation. In between, relaxation is established, probably through a "permeation" induced normal flow of matter through the cholesteric layers.

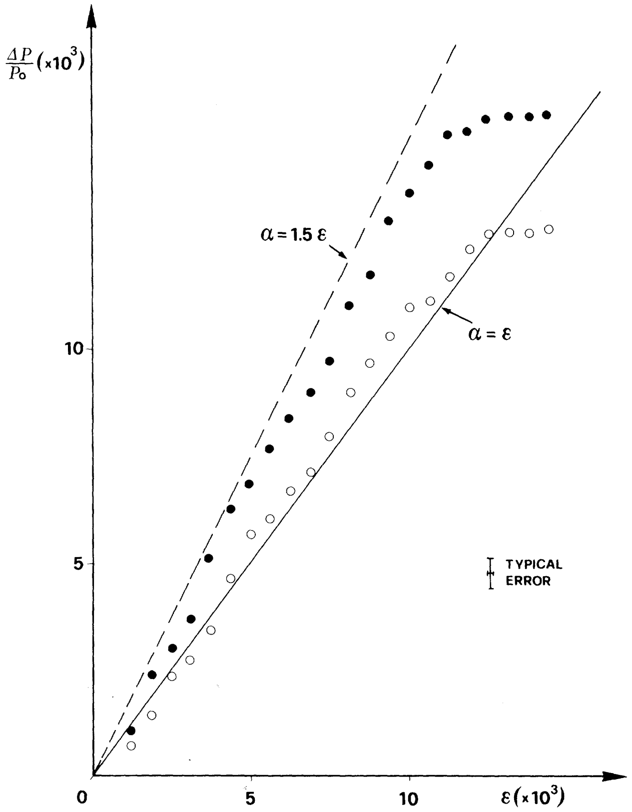

FIG. 3. Relative pitch variation $\alpha=\Delta p/p_0$ vs the external strain ϵ at long times (open circles) and 1 sec after the external strain application.

At shorter times, an unexpected inverted transient apparent strain is observed, which needs more quantitative analysis to be identified. A systematic effort is in progress to check the dependence of the observed effects on geometrical parameters (pitch, sample thickness, etc.), to ascertain the correspondence of our observations with the extension to cholesterics of the previous hydrodynamical models that describe flow in smectic liquid crystals.

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