Observation of Structural Instabilities in a Sheared Cholesteric

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Microscopic observations of the onset of structural instabilities in a cholesteric liquid crystal subjected to a static or a slowly periodic shear show that these instabilities are very similar to the buckling instabilities induced by a sudden dilation of the sample.

1. Introduction

Several authors [1–5] have studied the effect of a constant shear rate (steady state condition) on the planar structure of a cholesteric liquid crystal (CLC). In these cases the shearing is obtained by constant rotation of a rheogoniometer. The main conclusions are: 1) the CLC layers can slip freely, and a uniform helical axis tilt is produced at low shear rate. 2) a broken structure is induced in which the helix keeps a uniform orientation only inside small swarms. 3) the onset of hydrodynamical instabilities is observed. These conclusions have been criticized by some authors [6] because the rheogoniometer induces also a radial velocity gradient and because in the hydrodynamics backflow was ignored.

In our opinion the physical problem of the shearing of a CLC is not yet clarified because the nature of the induced instabilities is not well defined: structural and hydrodynamical instabilities can both be present and the cited experiments could favor one or the other.

In this paper a planar CLC is submitted to a static or slow linear shear deformation. This device simplifies the geometry and the discussion of the results.

We performed observations with an optical microscope and looked at the reflection spectra. Our conclusions are: 1) the layers slip, but not freely, 2) the helical axis tilts, 3) structural periodic instabilities take place before the onset of permanent deformations.

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2. Experimental

The cholesteric sample was a mixture of cholesteril chloride, nonanoate and oleylcarbonate, which shows a mesophase at room temperature and a small pitch dependence on temperature. The unperturbed pitch length is $P_0 = (4605 \pm 25)$ Å. The sample was placed between two glass plates (Fig. 1) of adjustable distance, the upper one being moved horizontally, either once or periodically by means of a hi-fi woofer loudspeaker. A good planar texture can be achieved by treating the glass surfaces with a silane agent. Vertical vibrations were prevented by teflon screws. The loudspeaker was excited by a

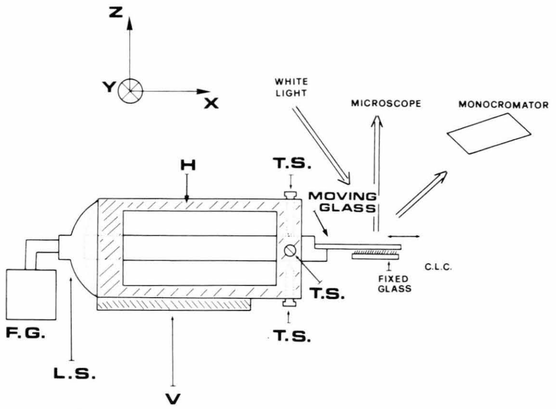

Fig. 1. Schematic drawing of the experimental apparatus: F. G. = Function generator driving the loudspeaker. V = Vertical movement of the whole support, made with micrometric skrew to regulate the moving glass position and thus the sample thickness. T. S. = Teflon skrews allowing a movement in y and z direction. H = Rigid holder. It is possible to rotate the loudspeaker position on the holder (rotation around x-axis) in order to regulate the planarity of the sample.

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waveform generator signal amplified by a 100 W amplifier. The system design was derived from one made in Paris to study the shear instabilities in nematics by the group at "Ecole normale de Physique ed Chimie" [7]. The displacements that can be obtained with our system range from 0 to 2 mm for static displacement and from 0 to 200 μm for 200 Hz excitation frequency. The displacements can easily be measured under the microscope even with periodic excitation by using a stroboscopic lamp of adjustable frequency. The sample parallelism is controlled to better than 10^{-3} rad, the sample thickness is (75 ± 5) μm.

We analyze the selective reflective band in the unperturbed and sheared states by the standard technique using a THR 1500 Jobin-Yvon monochromator. For studying the occurring tilts of the structure we performed microscopical observations and selective optical spectroscopy in the range of very small perturbations. We observed defect line displacements Δ in their dependence on the plate displacement S by viewing the liquid crystal/glass interface at a point where defect is visible at the glass surface.

3. Results

The results for static deformations are shown in Fig. 2, where one can see two types of linear behaviour: one for $0 < S < S_L$ and the other for $S_L < S < 30$ μm, where $S_L = 12$ μm. The observation that $\Delta < S$ indicates that the anchoring is not complete then the real shearing stress transmitted to the structure is not given by S but by Δ. The results for 100 and 200 Hz are shown in Figure 3. Two linear regimes are again observed, the first one having the same slope for both frequencies but its range decreasing with frequency. Figure 4 shows the position of the reflection peak λ_p vs. S for static deformation. In the first linear range of line displacements λ_p shifts toward shorter wave-lengths, as was observed earlier [8]. An undulated metastable structure is observed in the microscope when the shear is larger than a critical value which depends on the frequency of the applied disturbance. The buckling first appears around a defect or impurity, but for large enough deformations it is present everywhere. In any case it appears to be a local growth and not a propagation of deformations. We want to point out that what we show in Fig. 5 is not the real threshold for the onset of the instability, but the one where it becomes clearly visible. The reported points only give an upper limit and the general trend when the frequency increases. The undulations disappear in a few tens of ms if the frequency is low enough.

The transmitted light between crossed polarizers decreases abruptly when we go over $S \cong 30$ μm at 0.5 Hz, but then increases again in a time of $(50 \div 100)$ ms. When we further increase S ($S \sim 100$ μm), two relaxation times of the transmitted light appear: one of $(50 \div 100)$ ms and another of $(400 \div 500)$ ms. Presently it is not possible to get a direct relation between the data on the light transmission and the observations of the undulation because of the indetermination of the undulation threshold. At any rate we notice that the variation of the light transmission occurs at strain values close to those where the line displacements change slope and where the spectral behaviour changes.

The wavevector of undulation is parallel to the shearing direction; the wavelength is a bit larger than in the dilation case: (9 ± 2)μm here, (7 ± 2)μm in dilation, and it is independent of the frequency. This kind of deformation is fully reversible and it disappears when the mechanical field is switched off even at high frequency and with a recovering time increasing with the amplitude of S. For further increases of S a bidimensional deformation pattern substitutes the previous undimensional one which is progressively transformed into a focal conic pattern. Because of the depth resolution of the optical system (depending on the focal length and on the spatial resolution) deformations can not become visible before growing to some typical dimension: In our case we evaluated such a dimension to be $(0.5 \div 0.6)$μm. The focal conics network is stable and can partially relaxe after a few days. The initial planar configuration can be restored with a few large displacements at a very low frequency $\nu < 5$ Hz. The undulations and the focal conics pattern appear at shear displacements clearly decreasing with increasing frequency. The focal conics regime is observable only for frequencies larger than 1 or 2 Hz. The behaviour at low frequency and large displacements shows a progressive restoration of the planar texture.

For a further increase in the amplitude at $\nu > 5$ Hz some other effects are observed. We only list

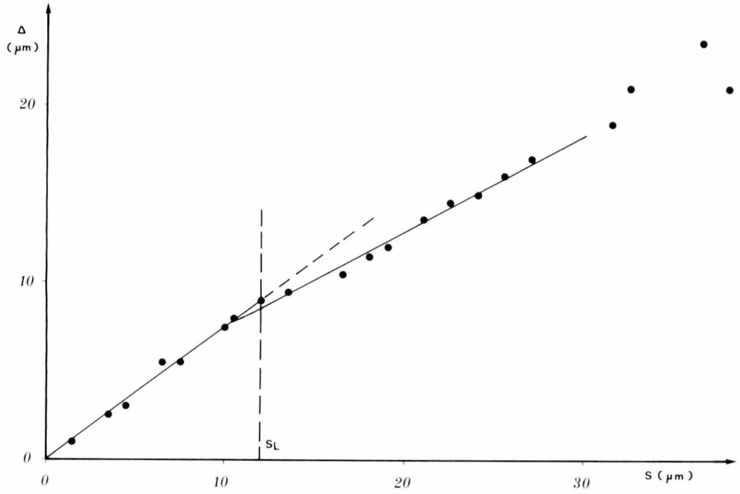
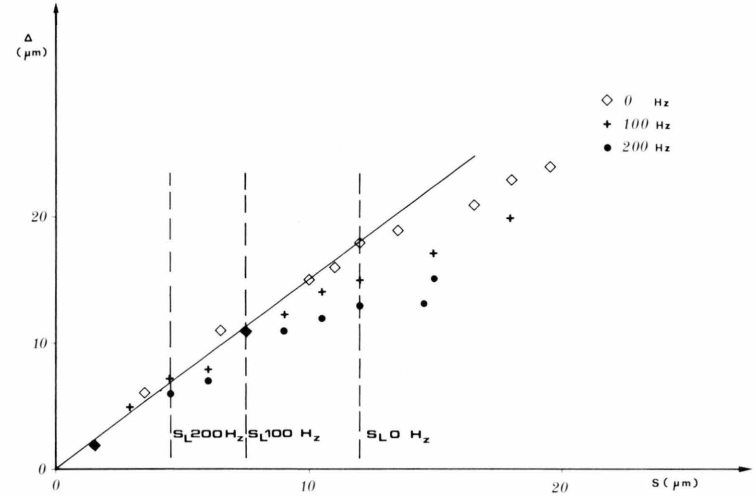


Fig. 2



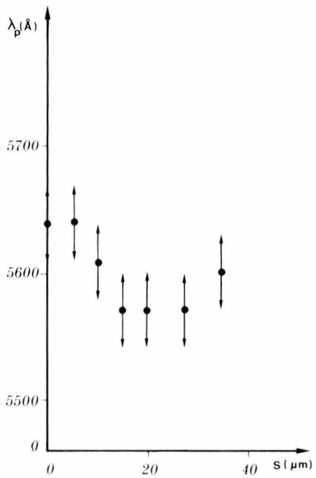


Fig. 4

Fig. 2. Displacement Δ of the attack point of a line defect at the upper plate versus the plate displacement S.

Fig. 3. The same as in Fig. 2 at different frequencies.

Fig. 4. Position of the maximum of the reflection peak λ_p versus the shear displacement S.

Fig. 5. Values of the shear for the optical appearance of the undulation and focal conics vs. the frequency of the driving signal.

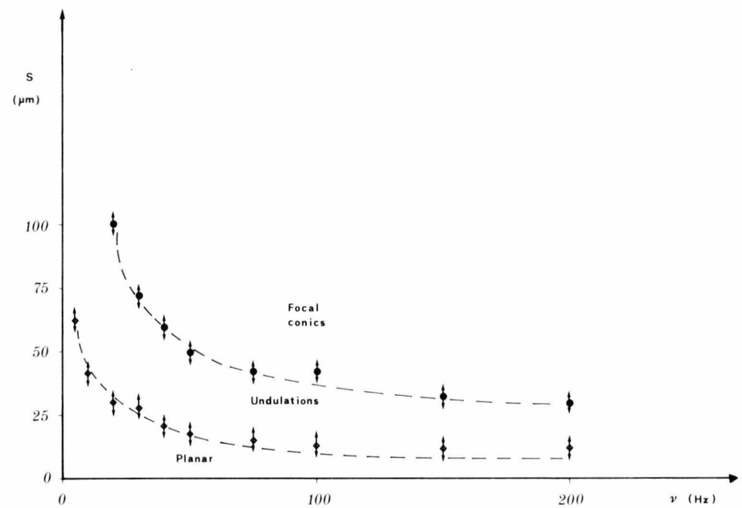


Fig. 5

them here and will report on them in detail in a future paper on the hydrodynamical properties. The sample assumes progressively an isotropic texture in which regular convective mouvements are visible. The propagation of singular waves is also visible and the transition into a blue phase can be achieved at the same time.

4. Discussion

The shape and the behaviour of the patterns observed are very close to those previously reported [9] for the buckling instabilities in smectics under mechanical or thermal dilations and to those we described earlier in cholesterics under dilation [10].

Since $\Delta > S$, we shall consider the measured Δ as the "true" displacement imposed on the liquid crystal. The existence of a linear response in displacement gives us the limits of existence of a non-dissipative and fully reversible process. The linear process involved in this region is a molecular tilt. In fact a molecular tilt involves a helical axis tilt β because of the long range chiral structure. The spectral behaviour allows us to obtain a rough indication of the imposed angle β_{IM}:

$$\beta_{\text{IM}} = \operatorname{arctg} \frac{\Delta}{d} \tag{1}$$

with d the distance of the plates. In this limit the reflection peak should shift towards shorter wavelengths if we consider that for a fixed internal incidence angle θ the reflection maximum is at rest

$$\lambda_{\text{p}} = P \cos \theta \tag{2}$$

and for helix tilted at an angle β_{IM}

$$\lambda_{\text{p}}' = P \cos (\beta_{\text{IM}} + \theta) \,. \tag{2'}$$

The measured shift of λ_{p} is at least one order of magnitude smaller than we would evaluate from (1), (2) and (2′). Of course the model of a rigid rod for the helical axis is too simple. However one can get the correct order of magnitude for the reflection shift by considering a wall effect imposing a spatial variation of the tilt angle and taking into account the reflectivity of each single cholesteric layer. We leave the discussion of these effects to a following paper [11]. The previous arguments allow us to consider our basic theoretical assumptions to be at least qualitatively correct. A shear induces a non uniform tilt of the structure and the tilt angle is roughly proportional to the shear itself. The undulations we observe under microscope can be interpretated as a consequence for higher strains. As we mentioned above, the accuracy of the actual system is not enough to determine the threshold values. Lastly we want to point out that, as in the dilation case, the undulations are followed by focal conics at large strain. In the smectic case, Clark [12] found a strain 1.7 times bigger than that for the buckling instabilities; here we find the same order of magnitude without any better precision because the resolution of the optical system is of the same order of magnitude of the depth or the deformation. On the other hand the accuracy of the pattern periodicity measurement is at least one order of magnitude better.

5. Conclusions

We have, for the first time, observed the formation of a periodic pattern in a cholesteric under a shear which we interpretate as the onset of buckling instabilities similar to those occurring under dilatation. The measurement of the undulation periodicity seems to support such a concept: one could try to justify qualitatively this result if there is some coupling between "cholesteric layers".

In fact a shear induces a tilt of a first cholesteric layer; then, if the layers are coupled, the tilt can be transmitted through the sample thickness over macroscopic dimension correspondent to several CLC layers, the CLC layers undergo an equivalent displacement because of the tilt. Above a critical tilt they undulate, or in an equivalent way buckling is produced when the shear displacement is greater than a critical value. The relationship between critical tilt and shear is not trivial because it should depend on many parameters such as, for example, the anchoring energy of the molecules on the surface and the length of the correlation between the CLC layers. If this picture of the CLC is true, we should consider it to be more like a solid than a nematic or a smectic. In fact, in smectics a solid-like elasticity is present normal to the layers which are uncorrelated. In the CLC a solid-like elasticity also exists and the "layers" are correlated. Following this idea we can not find buckling instabilities under shear in uncorrelated layered phases ($S_{\text{A}'}$, $S_{\text{C}'}$, some S_{B} [13], ...), but we should in some "more solid" correlated layered phases (S_{C}^*, some S_{B}, ...).

This idea is supported by the experiment of Kleman et al. [14] where they observed undulations also in a sheared smectic around some defects. We could consider that smectic layers near a defect (for example screw disinclinations) are strongly correlated. Light scattering measurements are currently underway to determine quantitatively the instability threshold in CLC and also to check the last hypothesis with different mesophases. Our results could be interesting and help to understand the mechanism for the onset of the hydrodynamical instabilities observed by several authors and by us for larger shear at high frequency because we can follow the shape of the deformation. These studies are underway as also a theoretical model for the observed effect.

Acknowledgements

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