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Field-Induced Cholesteric—Nematic Transition and Optical Bistability[†]

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The hysteresis of the field induced cholesteric \leftrightarrow nematic transition of mixtures of chiral compounds with nematics of positive dielectric anisotropy has been examined. The results show that the initial orientation of the sample is important and indicate that in those applications where a strong contrast is not essential, an initial homeotropic alignment gives a faster response and a better stability.

INTRODUCTION

The cholesteric-nematic transition (helix unwinding) induced by an electric (or magnetic) field perpendicular to the helical axis is well known. By gradually increasing the voltage applied to a cholesteric liquid crystal film of positive dielectric anisotropy, the transformation of the optically cloudy focal conic texture into a transparent nematic structure appears at a threshold field E_1 . When the field is removed, the reverse, nematic-cholesteric transition occurs, and its rate depends on the density of the residual disclinations, inhomogeneities, dust particles, etc., all of which act as nucleation centers. Relaxation times are a few ms (or less under certain conditions).

Greubel¹ first showed that by gradually decreasing the electric field the liquid crystal can remain in the nematic state up to a value E_2 lower than E_1 if the nucleation processes are slowed down. Lin-

[†]Paper presented at the 10th International Liquid Crystal Conference, York, 15th–21st July 1984.

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Hendel² has also observed this hysteresis and confirmed the possibility of two optical states being stable over a range of applied voltages $V_1 - V_2$ ($V_1 = E_1 \cdot d$, $V_2 = E_2 \cdot d$, d being the cholesteric thickness).

In this paper we describe the variation of the hysteresis range $V_1 - V_2$ with various parameters (cholesteric concentration, dielectric anisotropy, etc.), the stability of both optical states, the relaxation processes from the nematic to a cholesteric texture and the influence of the initial orientation of the sample on these various characteristics.

EXPERIMENTAL

A conventional experimental set-up was used. The liquid crystal film was held between two conducting glass plates separated by mylar spacers of thicknesses d between 6 and 18 μm . The various experiments were performed at room temperature with initial homogeneous alignment (a) obtained by rubbing the electrodes or a homeotropic alignment (b) obtained by deposition of a thin film of surfactant HTAB (hexadecyltrimethylammonium bromide). The liquid crystal samples used were mixtures of nematics (from E. Merck or Hoffmann-La Roche) with a positive dielectric anisotropy ϵ_a between 10.3 and 22.2 and a chiral substance (CB 15 from BDH); it must be noted that, due to the concentrations used, the pitch P is small compared to d , even for small cell thicknesses.

The experiments were carried out using a Leitz polarizing microscope: the light transmitted (for $\lambda = 0.59 \mu\text{m}$) by the liquid crystal impinging on the microscope photomultiplier, the anodic signal of which was registered as a function of the ac applied voltage on a X-Y recorder. The curve of transmission vs voltage was plotted point by point, the voltage being held constant for a sufficient time (30 s to 1 minute) to enable a quasi-stationary state to be attained. In this way the voltages V_1 and V_2 , which indicate the importance of the hysteresis effect can be determined for various concentrations c (between 5 and 15% by weight) of the chiral product and various values of the cell thickness d (or in other words as a function of the ratio d/p since the pitch P of the mixture is inversely proportional to the concentration c). These threshold determinations were made at a frequency of 100 Hz; preliminary experiments having shown that the values of V_1 and V_2 vary little with the frequency up to 5 kHz. The measurements of relaxation times between the nematic state and a given cholesteric texture were performed by observation of the anodic signal of the photomultiplier on an oscilloscope screen. Photographs

of the liquid crystal textures were taken during the relaxation processes.

RESULTS

1.) The general form of the relationship between the optical transmission and the applied voltage in a cycle of transformations, initial orientation \rightleftharpoons nematic state, is shown schematically in Figure 1 for homogeneous (a) and homeotropic (b) alignments. Several stages can be distinguished as the applied voltage is gradually changed. Some microscopic textures appearing during these transformations are illustrated in Figures 2a and 2b.

a) Homogeneous orientation. (Figures 1a and 2a). With no applied voltage the cell shows the classical Grandjean texture which is rapidly transformed at a voltage V_3 (between 5 and 10 volts depending on the values of the thickness d and of the concentration c) into a scattering focal conic texture; the latter state is maintained, as the voltage is increased, until the transition to the nematic state occurs: then the disclinations disappear and the optical transmission rises and saturates at the voltage V_1 . For gradually decreasing voltages the nematic structure remains up to the voltage V_2 and is stable for several hours for a fixed voltage between V_1 and V_2 ; in this range the stability of the state is related to the absence of nucleation centers (in particular residual disclination lines) and therefore to the time during which a voltage higher than V_1 has been applied. At the voltage V_2 a sudden relaxation process leads to a cloudy cholesteric state; for voltages lower than V_3 the cell returns to its original configuration more or less rapidly according to the concentration c .

b) Homeotropic orientation (Figures 1b and 2b). The behavior is rather similar to that indicated above, with the following differences. After the cell is filled, a structure which exhibits properties (optical activity, transmission) analogous to those of a Grandjean texture, is observed with no applied voltage; but in contrast to the preceding case this structure is unstable and is not formed again when the electric field is gradually decreased following the first cholesteric—nematic transition. The stability of the nematic state in the voltage range $V_1 - V_2$ is greater than for a homogeneous alignment and it can persist for several days in the absence of perturbations. In addition to this, the threshold V_1 (and therefore the range $V_1 - V_2$) are reduced at the second cycle of voltage variation.

2.) The voltages V_1 and V_2 deduced^{1,3,4} from the continuum theory

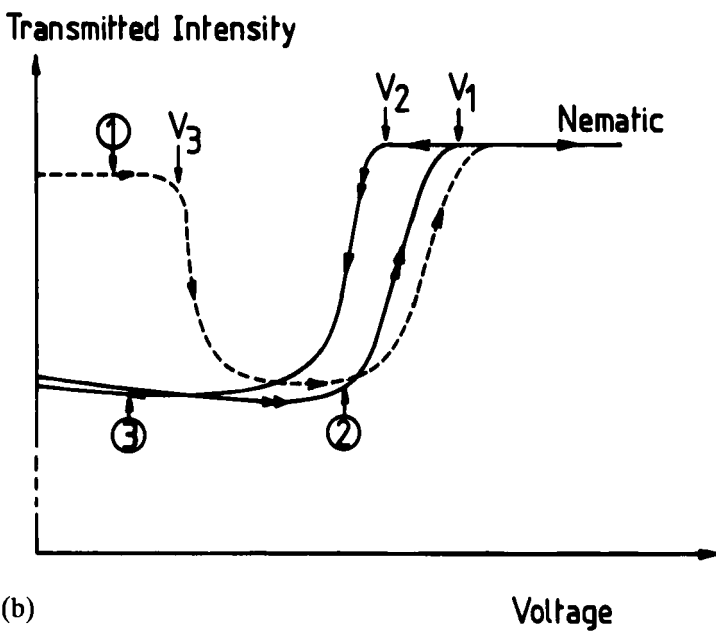
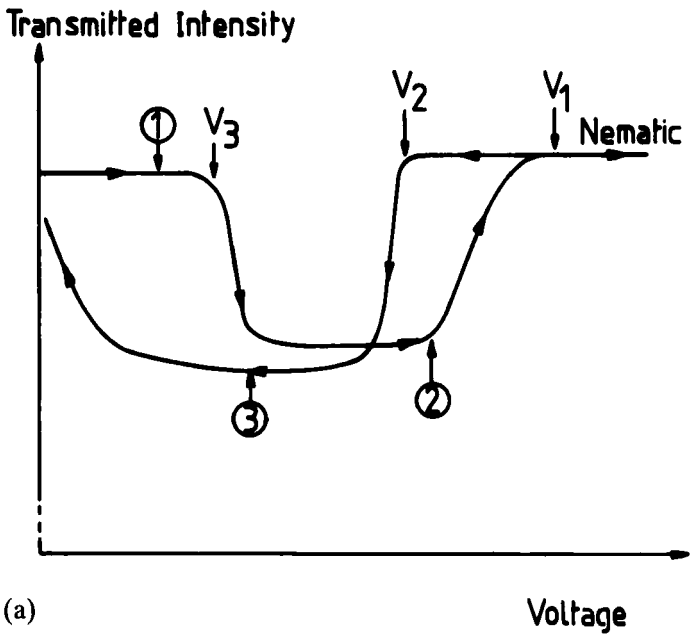
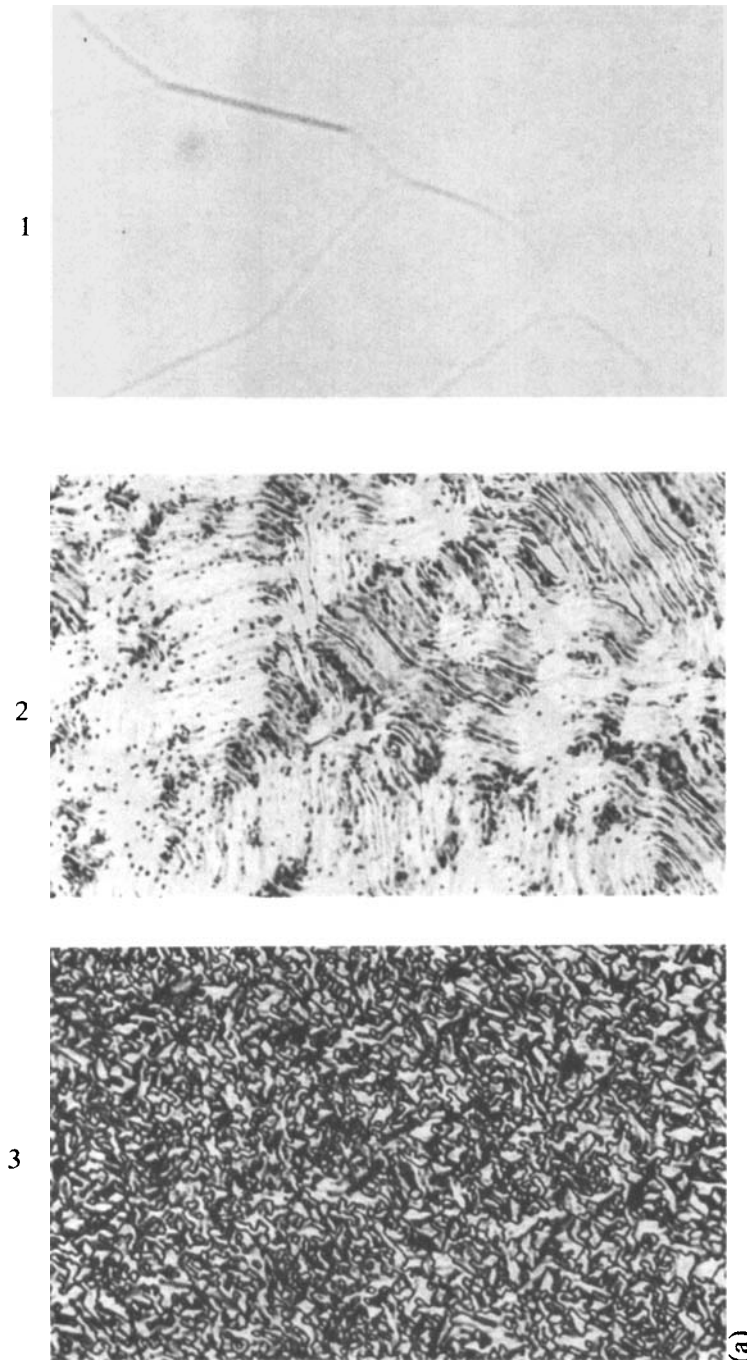


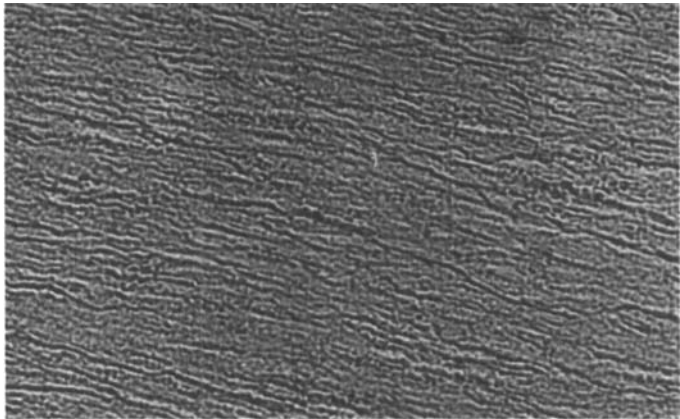
FIGURE 1 (a) Example of the transmitted intensity vs voltage in a homogeneous cell. (b) Example of the transmitted intensity vs voltage in a homeotropic cell.



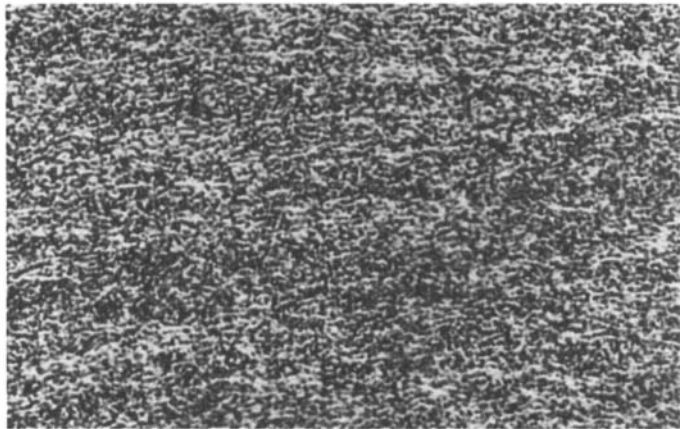
(a)

FIGURE 2 (a) Microscopic textures appearing for various voltages (see Figure 1a) in the homeotropic cell. (b) Microscopic textures appearing for various voltages (see Figure 1b) in the homeotropic cell.

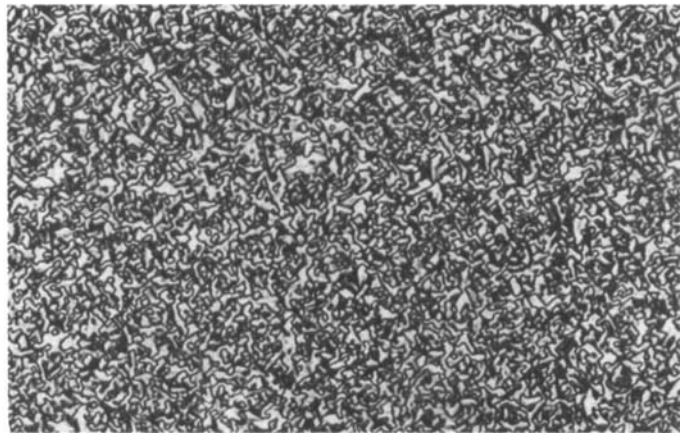
1



2



3



(b)

FIGURE 2 Continued

are given by the relations

$$\left\{ \begin{array}{l} V_1 = E_1 \cdot d = \pi^2 \frac{d}{P_0} \left[\frac{k_{22}}{\epsilon_0 \epsilon_a} \right]^{1/2} \\ V_2 = E_2 \cdot d = \frac{\pi}{(\epsilon_0 \epsilon_a)^{1/2}} \cdot \frac{d}{P_0} \left[\frac{4k_{22}^2}{k_{33}} - k_{33} \frac{P_0^2}{d^2} \right]^{1/2} \\ \qquad \qquad \qquad = \frac{\pi}{(\epsilon_0 \epsilon_a)^{1/2}} \left[\frac{4k_{22}^2}{k_{33}} \frac{d^2}{P_0^2} - k_{33} \right]^{1/2} \end{array} \right. \quad (1)$$

$$(2)$$

k_{22} and k_{33} being the elastic constants and P_0 the pitch for zero field. The ratio of both threshold fields

$$r = \frac{E_1}{E_2} = \frac{V_1}{V_2} = \pi \left[\frac{k_{22} k_{33}}{4k_{22}^2 - k_{33}^2 \frac{P_0^2}{d^2}} \right]^{1/2}$$

is independent of ϵ_a .

If $d \gg P$, r depends only on k_{33}/k_{22} and it is approximately equal to $\pi/2 (k_{33}/k_{22})^{1/2}$

According to relations (1) and (2) V_1 and also V_2 if $d \gg P_0$ must increase proportionally to the ratio d/P_0 or to the product c.d. In Figures 3 and 4 the experimental values of V_1 and V_2 are plotted vs. c.d. for three mixtures of CB15 and nematics of dielectric anisotropies $\epsilon_a = \pm 10.3, +19.2, +22.2$, the thickness being equal to 13 μm and the initial orientation homogeneous. These graphs show

—a linear variation of V_1 and V_2 in the range 5–15% of CB15 concentrations used

—a decrease (approximately as $\epsilon_a^{-1/2}$) of these thresholds for increasing dielectric anisotropy ϵ_a of the nematic; from a practical point of view it would be interesting to examine nematics of high dielectric anisotropy, but it must be noted that in this case the importance of hysteresis $V_1 - V_2$ for a given thickness d is reduced as predicted by the relations (1) and (2); it could be increased by using cells of greater thicknesses.

The influence of the initial orientation is not taken into account in the expressions (1) and (2). In Figure 5 the values of V_2 are plotted as a function of c.d for a mixture of 1132 TNC and CB15 ($c = 5\%$, $P_0 = 2.5 \mu\text{m}$) and for initial homogeneous and homeotropic align-

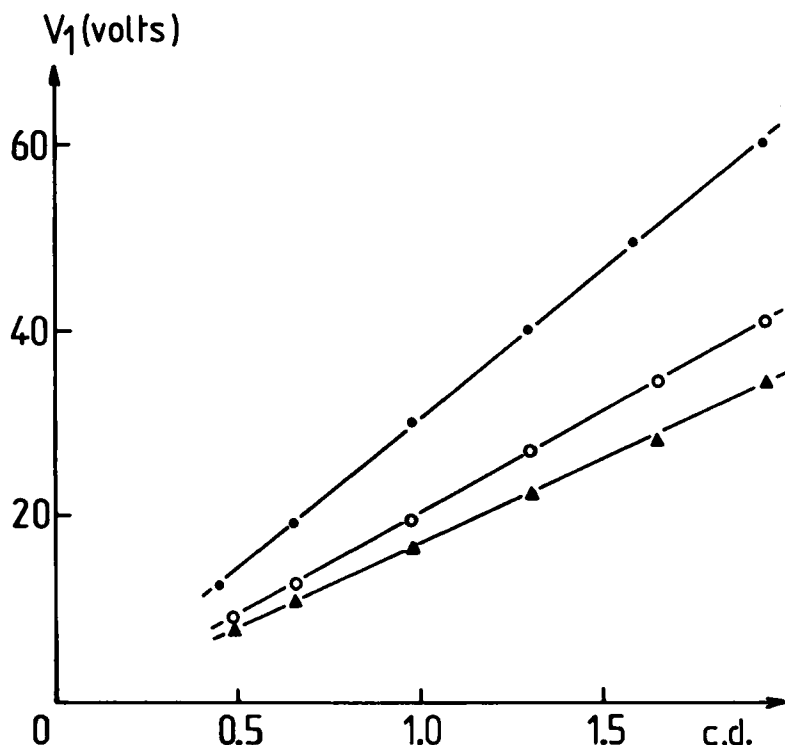


FIGURE 3 Experimental values of the threshold voltage V_1 vs c.d. for a homogeneous cell, $d = 13 \mu\text{m}$: ● 1132 TNC ($\epsilon_a = +10.3$) + CB15, ○ ROTN 403 ($\epsilon_a = +19.2$) + CB15, ▲ ROTN 132 ($\epsilon_a = 22.3$) + CB15

ments. It appears that the thresholds V_2 are, as expected,^{5,6} lower for the homeotropic than for the homogeneous condition.

It is possible to deduce values of elastic constants from these data: k_{22} can be obtained from the expression of V_1 (or from the slope of the straight line giving V_1 vs c.d) and k_{22}/k_{33} from the ratio V_1/V_2 , the pitch of each concentration being known. We have calculated these quantities for the three mixtures of Figures 3 and 4: it appears, for example, that the obtained values of constant k_{22} depend on the chiral product concentration and on the initial orientation; various authors^{7,8,9} have mentioned these difficulties related to different effects in particular the variation of the dielectric anisotropy by addition of a chiral product and non-uniform distortions during the cholesteric helix unwinding.

3.) We have also determined the turn-on (τ) and turn-off (τ') times when the electric field is applied or suppressed; τ and τ' are defined

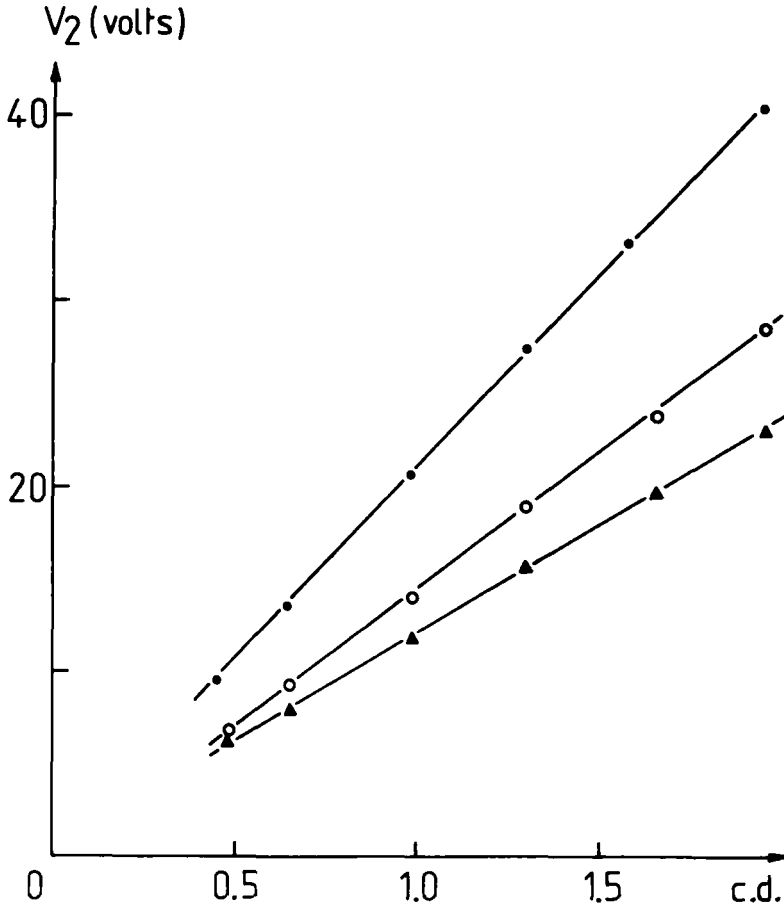


FIGURE 4 Experimental values of the threshold voltages V_2 vs c.d. for a homogeneous cell, $d = 13 \mu\text{m}$: ● 1132 ($\epsilon_a = +10.3$) + CB15, ○ ROTN 403 ($\epsilon_a = +19.2$) + CB15, ▲ ROTN 132 ($\epsilon_a = +22.3$) + CB15

as the total times corresponding to the transition from the initial state into the nematic texture and to the reverse.

i) For a sudden increase of voltage from zero to $V(V > V_1)$ the time τ rises rapidly for decreasing fields as observed for the pure twisted nematic,¹⁰ but the presence of a chiral product leads to turn-on times much shorter than for the pure nematic: for example with a cell TNC 1132 + 5% CB15 ($d = 6 \mu\text{m}$) τ varies from 8 ms to 35 ms for voltages from 60 to 30 volts. Small differences in the values of τ are found for the two initial alignments, the homeotropic orientation corresponding to the shorter times.

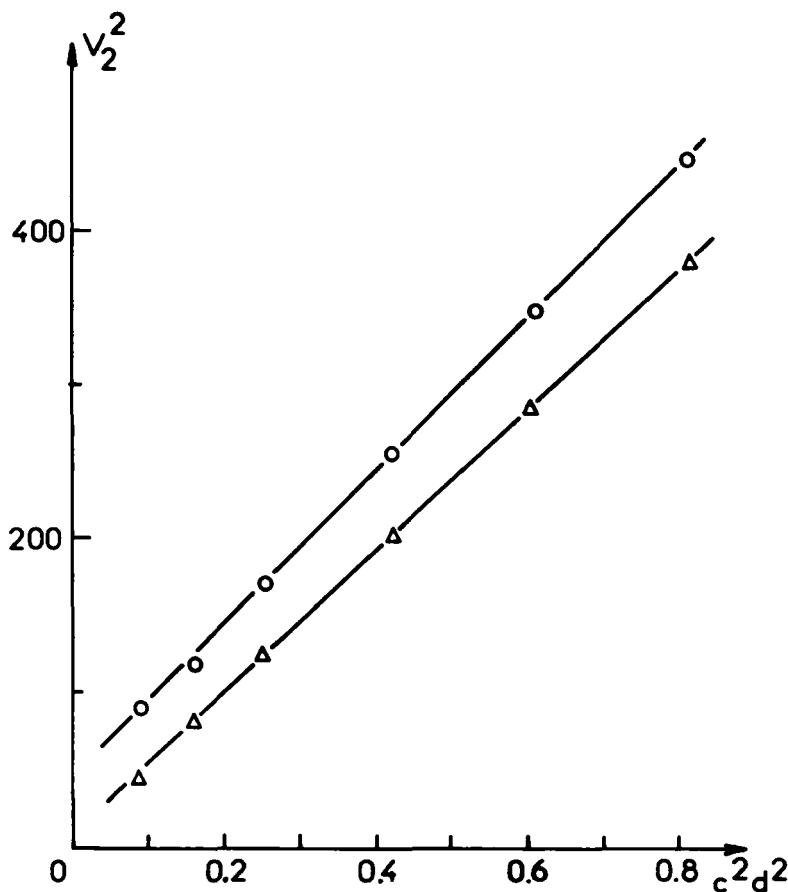


FIGURE 5 Experimental values of the threshold voltages V_2^2 vs $c^2 d^2$ for cells of 1132 TNC + CB15 (5%): \circ homogeneous, \triangle homeotropic

ii) When the applied voltage within the range giving a nematic state ($V > V_1$) is suddenly suppressed, the turn-off time τ' , which is practically independent of the initial orientation and of the voltage V , decreases drastically for higher chiral concentrations c : with the cell described in the preceding paragraph, τ' is equal to 90 ms for $c = 5\%$ and to 7 ms for $c = 15\%$. The influence of the ratio d/P_0 and of the dielectric anisotropy ϵ_a on τ' is shown in Figure 6 where τ'^{-1} is plotted vs $c^2 d^2$ ($\propto d^2/P_0^2$) for $\epsilon_a = 10.3$, $d = 6, 10, 13 \mu\text{m}$ and for $\epsilon_a = 22.2$ $d = 13 \mu\text{m}$; it appears that τ'^{-1} varies linearly with d^2/P_0^2 .

However, if the voltage is reduced to a value v ($0 < v < V_2$) the processes are different; the characteristic time τ'_w which corresponds

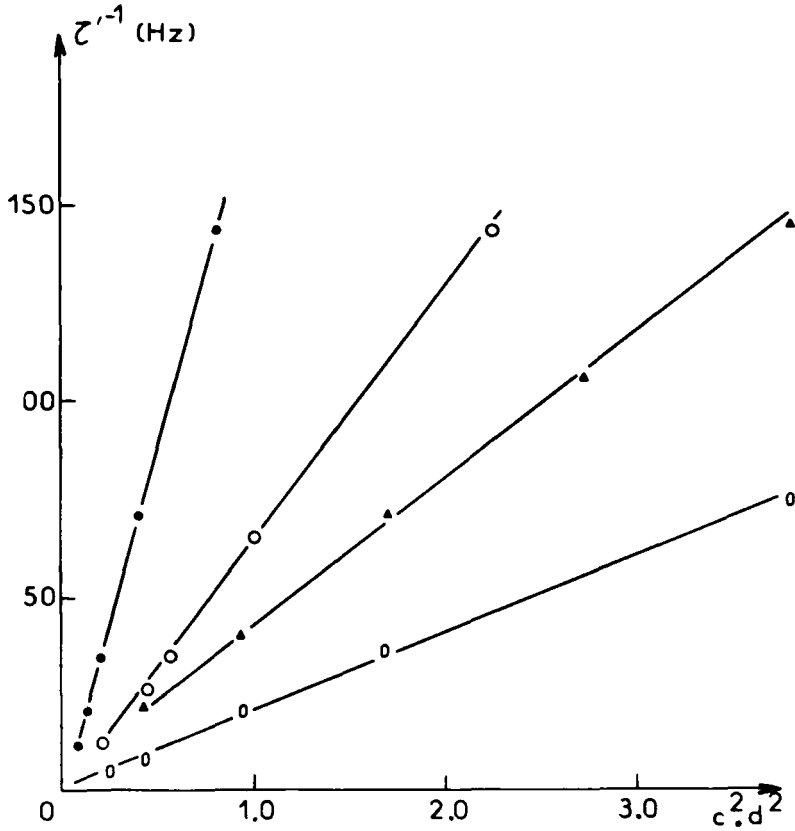


FIGURE 6 Reciprocal of the relaxation times τ' vs $c^2 d^2$ for different mixtures and thicknesses: $\epsilon_a = 10.3$; ● $d = 6 \mu\text{m}$; ○ $d = 10 \mu\text{m}$; ▲ $d = 13 \mu\text{m}$; $\epsilon_a = 22.3$; ○ $d = 13 \mu\text{m}$

to the appearance of a focal conic structure (or a 'winding' process) is equal to τ' for $\nu < V_3$; if $\nu > V_3$ τ'_w increases with ν , as observed by Gerber¹¹ with mixtures of nematic and CB15 for low concentrations. This variation depends on the initial orientation as shown in Figure 7 giving $(\tau'_w)^{-1}$ vs ν ; it can be seen that τ'_w is shorter for the homeotropic alignment than for the homogeneous one.

4.) Finally we have considered the possibility of utilizing this hysteresis in an optical bistable device. If the cell is maintained in the optically cloudy focal conic texture by applying a suitable voltage V_0 (between V_1 and V_2) it can be turned into the transparent nematic state by a voltage pulse and will remain in this new state in the absence of nucleation centers; its stability is related, as described above, to the duration and the amplitude of the pulse which must be sufficient

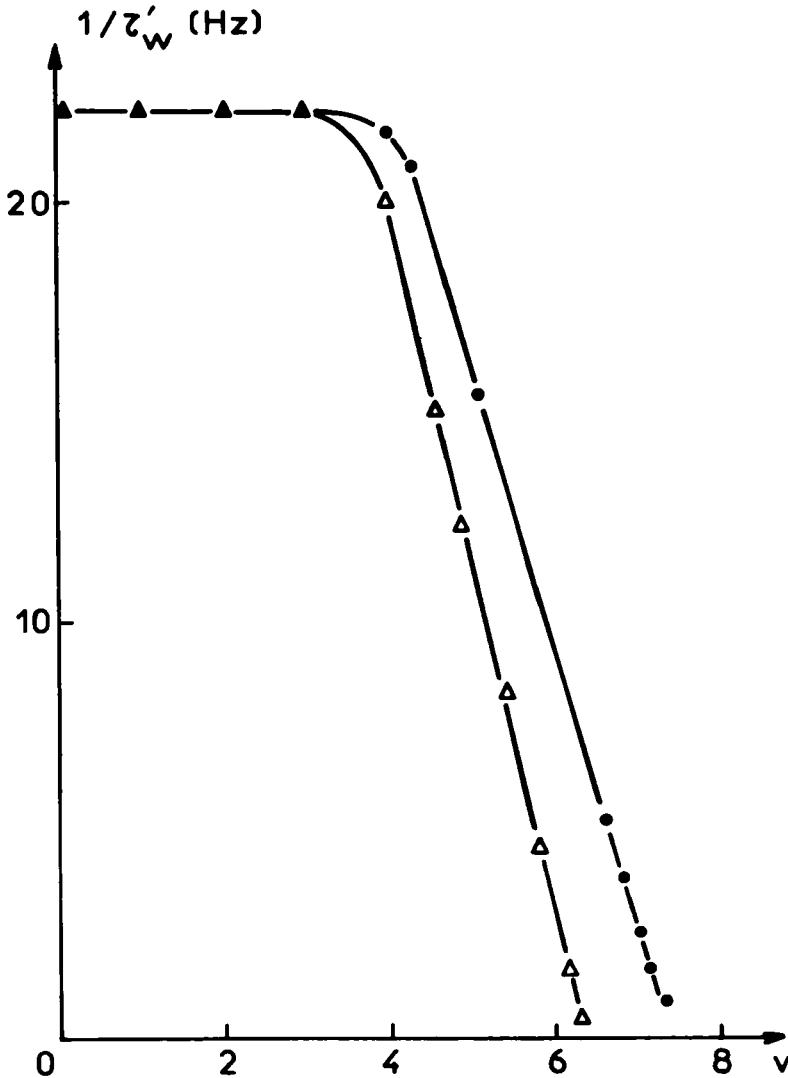


FIGURE 7 Reciprocal of τ'_w ('winding' time: from nematic into focal conic) vs voltage v , for cells of 1132 TNC + CB15 (5%), $d = 8 \mu\text{m}$: Δ homogeneous, \bullet homeotropic.

(for example ≈ 10 ms and 20 volts above V_1) to remove any causes of nucleation. A negative pulse of amplitude higher than $V_0 - V_2$ causes the cell to revert to the focal conic texture.

CONCLUSION

We have been able to determine the characteristics of the hysteresis in mixtures of cholesterics with nematics of positive dielectric anisotropy, when they are submitted to electric fields, as well as the times that characterize the cholesteric \rightleftharpoons nematic transitions.

Our results show the influence of the initial orientation, and stress that it is advisable, for those applications where strong contrast is not essential, to use a homeotropic alignment for faster response and better stability.

Acknowledgments

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