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Dynamics of Molecular Orientation, during the Cholesteric → Nematic Transition†

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Received October 2, 1972

Abstract—We present a study of the dynamics of transition from the cholesteric to the nematic state and reverse. A theoretical interpretation is given according to the continuum theory, which leads to the determination of the following ratios:

$$\frac{K_{22}}{\gamma_1}$$
, $\frac{K_{22}}{\Delta \chi}$ and $\frac{\gamma_1}{\Delta \chi}$

A good agreement is found with previous measurements.

1. Introduction

Our aim was not, in the present work, to build a display device exhibiting very quick responses to external stresses as in Refs. I and 2, but to give a kinetic description of the transition cholesteric \rightleftharpoons nematic, induced by switching on (off) a magnetic field. So our attention was drawn by the conditions leading to a "monocrystalline" evolution in both phases (i.e., no disclinations and complex textures). At least one boundary had to let the molecules rotate freely, because their angle with a fixed axis (let's say the direction of the field) had to pass continuously (and homogeneously in a plane containing the direction of the field and perpendicular to the helical axis) from $a\pi(a>1)$ to nearly zero, which is impossible with two rubbed surfaces. For those quite simple reasons we decided to study the behaviour of a drop in the Grandjean planar structure.

[†] Presented at the Fourth International Liquid Crystal Conference, Kent State University, August 21-25, 1972.

2. Experimental Set Up

We can see on Fig. 1 the experimental device allowing the study of the transition. The drop is placed on a rubbed glass surface, itself inserted between the poles of a permanent magnet. We get its image on a screen (\times 100) and take the intensity of light at one point of the pattern, with a photomultiplier tube connected to a PDP 8 computer.

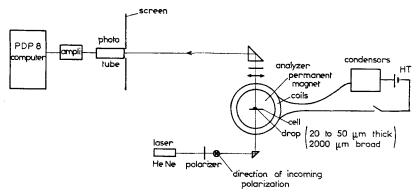


Figure 1. Experimental set up.

The shortest time for a measurement is 2.10^{-4} sec, but we can make it much longer and take the mean value. A curve is composed of 4,000 points, the overall duration of the evolution determines the time of a measurement.

The intensity of the magnetic field can vary from 0 to 11,000 Gauss (reciprocally 11,000 G to 0) and the change is imposed in a few 10^{-2} seconds (in the less favourable cases it can reach 10^{-1} second).

The intensity value is given by a Hall probe and known with 3% of uncertainty.

The observation is made between crossed polarizers, the incident polarization of light being parallel to the field and to the rubbing direction. In the Mauguin limit⁽³⁾ the polarization of the outgoing beam gives the direction of the optical axis at the upper surface. For that reason, we investigated the behaviour of mixtures of MBBA and cholesteryl oleate the pitch of which is in the range of 10 μ m.

3. Typical Behaviour at Transition

A. CHOLESTERIC → NEMATIC

Before the field is applied, the drop (~ 2 mm diameter and 20 to 50 μ m thick) exhibits between crossed polarizers concentric dark circles which correspond to the Grandjean planar structure (Fig. 2). When it is switched on shadowed regions first appear followed by dark circular threads which begin to move toward the top of the drop (Fig. 3). In fact, the threads are the intersection with the

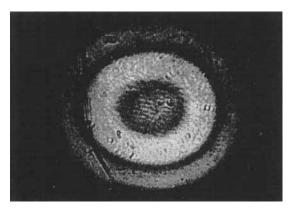


Figure 2. Drop on the Grandjean planar structure ($p_0 = 9.7 \,\mu\text{m}$). (Crossed Polarizer; Polarization parallel to the field.)

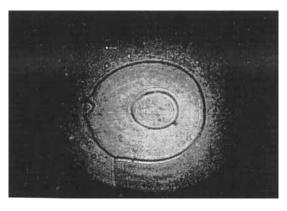
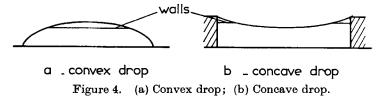


Figure 3. "Thread" moving toward the centre of the drop. (Crossed Polarizer; Polarization parallel to the field.)

surface of π walls which are formed in the bulk of the drop. We can make sure of it, in two ways:

i) The direction of the first movement of the wall is essentially governed by the change in energy due to surface variation. Then if we study a "concave" drop (Fig. 4b), the wall moves in a direction which minimizes its energy, that is they tend to leave the centre, contrary to the case of the convex drop (Fig. 4a). This is well verified experimentally. If we were dealing with singularities like those described in Rault's paper (4) the movement would not be the same.

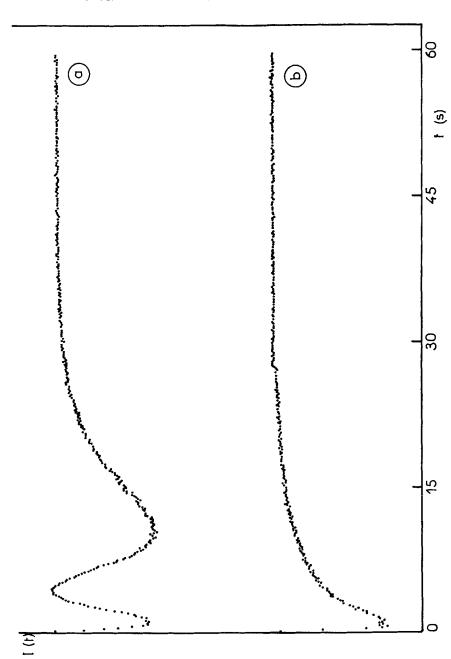


ii) We can also prove it by letting the structure relax to the cholesteric state, in and out of the presence of a wall. The comparison of the two curves clearly shows that there is a π difference between the director direction at the free surface with and without the wall (Fig. 5).

During the cholesteric \rightarrow nematic transition, the migration takes rather a long time, the whole process has a duration of the order of ten or twenty minutes. During that time the director angle at the surface has a fixed value (Fig. 6), except when a wall is expelled. In the final state, the director direction at the surface has still the same value which is not that of the field: the extinction is not obtained (Fig. 7) except in the limiting case $H \gg H_C$. In the bulk, the nematic order is reached, but the sample bears a memory of the cholesteric order at the free surface.

We can sum up the process in the following way: there is a rapid creation of π walls, which slowly move toward the free surface.

N.B.—If the applied field is much higher than the critical one $[H_C = \pi^2/2\sqrt{(K_{22}/\Delta\chi)}\ 1/p_0]\ K_{22}$ twist elastic constant, $\Delta\chi$ magnetic anisotropy, p_0 period of the structure, then under certain circumstances disclination lines appear and relax the structure quickly. We are not dealing with such a process.



(a) Relaxation to the cholesteric state, when there remains no "thread" in the drop, before switching off (b) Relaxation to the cholesteric state, when there remains one thread in the same drop, before switching off Figure 5. the field. (the field.

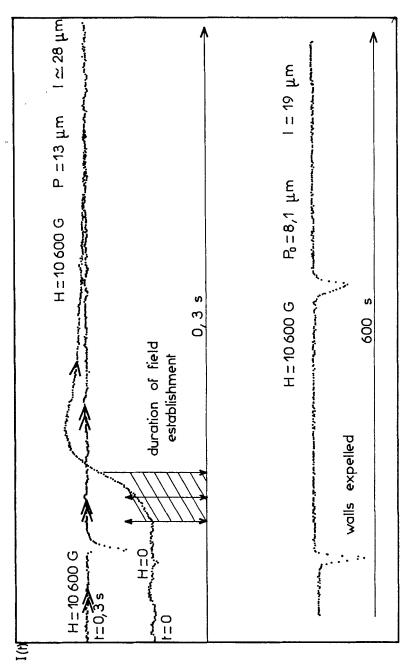


Figure 6. Photomultiplier response, after the field is applied. Note the constancy of the intensity, excepted when a wall is expelled.

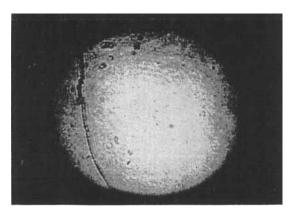


Figure 7. Drop after the transition.

B. Nematic → Cholesteric

When the field is suddenly switched off, the Grandjean planar structure is restored in a few seconds. The director clearly rotates at the free surface, till the initial pattern is reached (Fig. 5). The Grandjean planes appear at the centre of the drop (Fig. 8) before taking their initial place.

With such a geometry, no complex texture appears as in Ref. 1. All those features show that it is possible to describe the transition simply with the aid of the continuum theory.

4. Theoretical Interpretation

Using Ericksen Leslie continuum theory (5,6) we have to solve the following equations, adapted to our problem:

$$\begin{bmatrix} \boldsymbol{V}_{i,\,i} = \boldsymbol{0} \\ \boldsymbol{O} = \boldsymbol{g}_i + \boldsymbol{\pi}_{ji,\,j} \end{bmatrix} \boldsymbol{\sigma}_{ji,\,j} = \rho \, \frac{\mathrm{d}\,\boldsymbol{V}_i}{\mathrm{d}t}$$

where:

 V_i is the velocity of the fluid

 σ_{ji} the stress tensor

 g_i the intrinsic director body force

 π_{ji} the director stress tensor

i denotes the derivation with respect to the space coordinate. The Einstein summation convention is adopted.

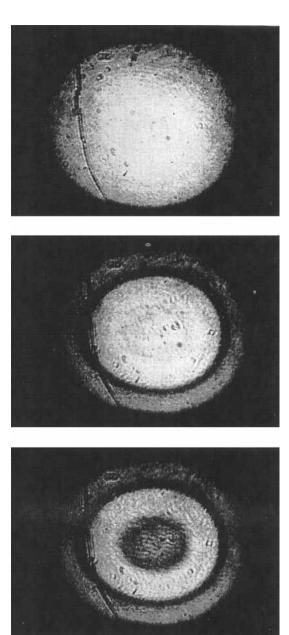


Figure 8. Restoration of the Grandjean planar structure. (Crossed Polarizer; Polarization parallel to the field.)

Let us consider an infinite cholesteric film (in the x, y directions) in the Grandjean planar structure, the helical axis being borne by the z direction; let us call \mathbf{d} the director, so it has the following components:

$$\begin{cases} dx = \cos \theta \\ dy = \sin \theta \\ dz = 0 \end{cases}$$

Due to the infinity in the x, y directions, θ is only a function of z and the time t. Using Leslie's development for the non equilibrium terms, and Frank elastic theory,⁽⁷⁾ the movement equations are easily derived: one can show:

- i) there is a consistent solution, uncoupled with hydrodynamical movement (Vx = Vy = Vz = 0 everywhere in the film).
 - ii) the pressure in one point is given by the following equality:

$$P = P_0 + \left(\frac{\partial \theta}{\partial z}\right) \left[K_2 - K_{22} \left(\frac{\partial \theta}{\partial z}\right)\right]$$

where P_0 is the external pressure, K_{22} and K_2 are elastic moduli, defined by Frank.⁽⁷⁾ P is expected not to be significantly different from P_0 .

iii) at the rubbed surface θ is fixed in the field direction

$$\theta(z=0,t) = \frac{\pi}{2} \tag{1}$$

but at the free surface, if we assume that the director rotates freely and is parallel to it:

 $\pi_{jk} \nu_j = 0$ (v perpendicular to the surface)

we get:

$$\frac{\partial \theta}{\partial z}(z=l,t) = \frac{\pi}{p_0} \tag{2}$$

Boundary condition already used by Leslie (6)

iv) the movement equation of the director is:

$$\gamma_1 \frac{\partial \theta}{\partial t} - K_{22} \frac{\partial^2 \theta}{\partial z^2} = \frac{\Delta \chi H^2}{2} \sin 2\theta \tag{3}$$

H = applied field

 $\gamma_1 = -\lambda_1 = \text{twist viscosity constant.}$

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This relation is typically non linear; a unique solution exists, if we add the initial condition:

$$\theta(z, t = 0) = f(z) \tag{4}$$

A. RELAXATION TO THE CHOLESTERIC STATE

We first deal with the relaxation process because of its simplicity. The equation reduces to:

$$\gamma_1 \frac{\partial \theta}{\partial t} - K_{22} \frac{\partial^2 \theta}{\partial z^2} = 0.$$

The initial value of the function is the solution of Eq. (3) with $\partial \theta/\partial t = 0$, and the conditions of Eqs. (1) and (2); we get:

$$\theta(z, t = 0) = \frac{\pi}{2} + \mu \exp\left(\frac{z - l}{\xi_i}\right)$$

with:

$$\mu = \frac{p_0}{\pi \xi_i} - \sqrt{\frac{{p_0}^2}{\pi^2 {\xi_i}^2} - 1} \qquad \qquad \xi_i = \sqrt{\frac{K_{22}}{\varDelta \chi}} \cdot \frac{1}{H_i} \label{eq:mu_energy}$$

 ξ_i is the coherence length already defined by De Gennes,⁽⁸⁾ H_i is the initial value of the field.

The memory of the cholesteric state remains in a thickness corresponding to that coherence length. Everywhere else, $\exp(z - l/\xi_i) \simeq 0$, and $\theta \simeq \pi/2$. Note that for $H < H_C$, the pattern is not strongly disturbed till a critical field

$$H_{CI} = \pi \sqrt{\frac{K_{22}}{\varDelta \chi}} \frac{1}{p_0}$$

is reached. Above this value the cholesteric drop can be considered as a succession of π walls, which are stable as long as $H < H_C$.

Let us write down:

$$\alpha_1 = \theta - \frac{\pi}{2} - \frac{\pi z}{p_0}$$

The new function is defined in the domain $z \in [0, l]$ and $t \in [0, \infty]$; we can continue it outside the domain in the following way:

$$\begin{bmatrix} \alpha_1(-z) = -\alpha_1(z) \\ \alpha_1(l+z) = \alpha_1(l-z) \\ \alpha_1(z+4l) = \alpha_1(z) \end{bmatrix}$$

The solution (ensuring the good boundary conditions) is then written as the Fourier expansion:

$$\alpha_1 = \sum_{n=0}^{\infty} a_n \exp \left[-\frac{K_{22} \pi^2}{\gamma_1 l^2} (n + \frac{1}{2})^2 t \right] \sin \left[(n + \frac{1}{2}) \frac{\pi z}{l} \right]$$

with:

$$a_n = \frac{2}{l} \int_0^l \left[\theta_0(z, t = 0) - \frac{\pi}{2} - \frac{\pi z}{p_0} \right] \sin \left[(n + \frac{1}{2}) \frac{\pi z}{l} \right] \mathrm{d}z$$

For the sake of simplicity we can neglect the influence of the coherence length, which is only important in the very beginning of the relaxation. Coming back to the θ expression, we get:

$$\theta = \frac{\pi}{2} + \frac{\pi z}{p_0} - \sum_{0}^{\infty} (-1)^n \frac{2l \exp\{-[K_{22}\pi^2(n+\frac{1}{2})^2t]/\gamma_1 l^2\}}{p_0(n+\frac{1}{2})^2\pi} \sin\left[(n+\frac{1}{2})\frac{\pi z}{l}\right]$$

The experimental curve is given by:

$$I(t) = \cos^2 \left(\theta(z = l, t)\right)$$

that is:

$$I(t) = \sin^2 \left\{ \frac{\pi l}{p_0} - \sum_{0}^{\infty} \frac{2l}{\pi p_0 (n + \frac{1}{2})^2} \exp \left[\frac{-K_{22} \pi^2 (n + \frac{1}{2})^2 t}{\gamma_1 l^2} \right] \right\}$$

The consistency of this interpretation, can be easily checked.

The pitch in zero field is measured by the Canowedge method. The thickness of the drop is determined by counting the interference fringes exhibited by the drop in suitable incoming polarization and "infinite" applied field (Fig. 9).

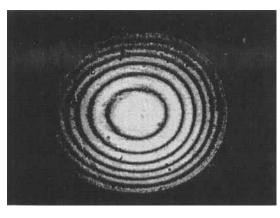
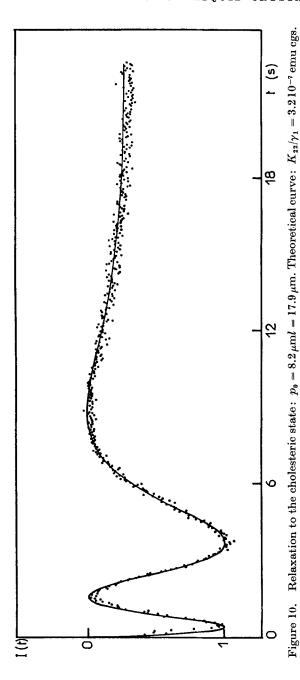
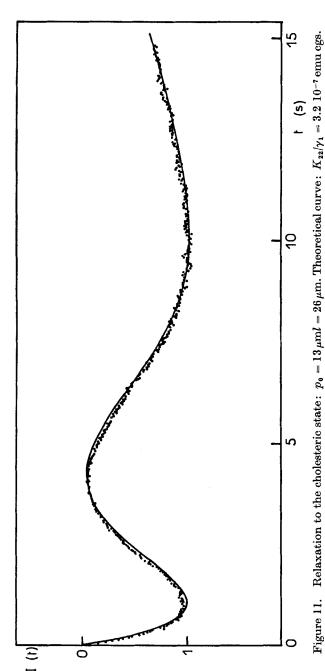


Figure 9. Interference fringes. (Crossed Polarizer; Polarization at $\pi/4$ from the field)—(Applied field: 10,600 gauss).





Figures 10 and 11 show the very good agreement between theory and experiment. The theoretical curve is calculated by the computer and traced on a scale directly comparable to the experimental one.

The best fit is obtained with the ratio:

$$\frac{K_{22}}{\gamma_1} = 3.2 \pm 0.1 \ 10^{-7} \, \text{emu cgs}$$

Other curves obtained by changing the thickness (in the range $l < 3p_0$, necessary to observe a simple relaxation) give a very satisfactory agreement between theory and experiment, with the same ratio value.

From other techniques, we know the K_{22} and γ_1 values^(9,10,11) of MBBA. The small amount of cholesterol derivatives added does not change it significantly. Our present determination is consistent with the values reported previously. We think that the study of the relaxation provides a simple and rapid tool to determine such a ratio. It is also a good test to check the validity of the boundary condition at the upper surface

$$\frac{\partial \theta}{\partial z} = \frac{\pi}{p_0}$$

B. Transition Cholesteric → Nematic

As we have already pointed out, the process is essentially governed by the migration of π walls in the sample. So this problem is very near to that studied theoretically by De Gennes⁽¹²⁾ and experimentally by L. Leger⁽¹³⁾, with two further complications due to the fact that the boundary condition at the free surface acts itself like a wall, and that a drop is not infinitly broad.

Passing at the functional limit ($\theta = \pi Y_s + \pi/2$, where Y_s is the Heaviside step function located at the position s of the wall) one can show that the solution can be split into two parts: a static (or elastic) one which gives the "shape" of the wall, and a second one which gives its movement.

Such a presentation cannot allow us easily to take account of the surface variation of walls, but we keep as a result, that "static" forms move, in the drop. We can now use a method quite similar to the one already described by De Gennes. (12) Let us consider the case of the reference: a single wall in a thick sample $(l \gg \xi)$. The

free energy of the wall takes the form:

$$F = \frac{S(s)}{2} \int_0^\infty \mathrm{d}z \left\{ K_{22} \left(\frac{\partial \alpha}{\partial z} \right)^2 - 2 q_0 \! \left(\frac{\partial \alpha}{\partial z} \right) + \varDelta \chi \; H^2 \; \mathrm{sin^2} \; \alpha \right\}$$

$$\alpha = \theta - \frac{\pi}{2}.$$

In this expression F is developed around a nematic unperturbed solution, which corresponds to the stable one when the wall is expelled, and must thus be taken into account when there are surface variations. The surface S(s) occupied by the wall can be taken before integrating because the energy is strongly located around s. After some calculations we get:

$$F = \frac{1}{2} \Delta \chi H^2 \left[4\xi + 8\xi \lambda^2 - 2\xi^2 \frac{\pi^2}{p_0} + 0(\lambda^3) \right] S(s)$$

with $\lambda = \exp(-s/\xi)$.

To obtain this expression, we only need to give a "static" form of the wall, namely:

$$tg\left(\frac{\alpha^{+}}{2}\right) = \exp\left(\frac{z-s}{\xi}\right)$$
$$tg\left(\frac{\alpha^{-}}{2}\right) = \exp\left(\frac{-z-s}{\xi}\right)$$

This form is identical to the one obtained by De Gennes for a nematic. The α^- part ensures the boundary condition $\alpha(z=0)=0$ and is the image of the wall described by α^+ .

Then:

$$-rac{\mathrm{d}F}{\mathrm{d}s}=S(s)8arDelta\chi H^2\lambda^2-rac{1}{2}rac{\mathrm{d}S}{\mathrm{d}s}\,arDelta\chi H^2igg[4\xi+8\xi\lambda^2-2\xi^2rac{\pi^2}{p_0}igg]$$

The dissipated power is identical to that in the nematic phase:

$$P = S(s)\gamma_1 \int_0^t \left(\frac{\partial \alpha}{\partial t}\right)^2 \mathrm{d}z \simeq \frac{2\gamma_1}{\xi} \frac{V^2}{\xi} S(s)$$

where V = ds/dt is the velocity of the wall.

This system being in a field independent of time, the speed of free energy variation is equal to the dissipated power. Equating the two, we find the same velocity as given by De Gennes in the nematic case, apart from the term due to surface variations:

$$V = 8\omega_0 \xi \left\{ \exp\left[-2s/\xi\right] - \frac{\xi}{8} \left(2 - \frac{\pi^2 \xi}{p_0}\right) \frac{d}{\mathrm{d}s} \log\left(S\right) \right\}$$

In fact in the drop, more than one wall is present. If we note that the image method gives the interaction between two walls, the generalization is straight forward (Fig. 12).

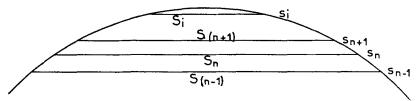


Figure 12. Location of the walls.

$$\begin{split} Vn &= 8\omega_0 \xi \left\{ \exp\left[-(s_n - s_{n-1})/\xi - \exp\left[-(s_{n+1} - s_n)/\xi\right]\right] \right. \\ &\times \frac{S_{n+1}}{S_n} - \frac{\xi}{8} \left(2 - \frac{\xi \pi^2}{p_0}\right) \frac{d}{\mathrm{d}s} \log S \right\} \end{split}$$

By writing such an equality we neglect border effects which are expected to be small.

At the free surface, we must satisfy the boundary condition:

$$\frac{\partial \alpha}{\partial z} = \frac{\pi}{p_0}$$

Let us call s_i the position of the wall located just below the surface, α_i the angular function corresponding, and α_t the function corresponding to the part of wall at the surface itself.

$$tg\frac{\alpha_i}{2} = \exp\left(\frac{z-s_i}{\xi}\right)$$
 and $tg\frac{\alpha_t}{2} = \mu \exp\left(\frac{z-l}{\xi}\right)$

With a good degree of approximation: $\alpha = \alpha_i + \alpha_t$ (in the domain $z \sim l$).

Then:

$$\frac{\partial \alpha}{\partial z} = \frac{\partial \alpha_i}{\partial z} + \frac{\partial \alpha_t}{\partial z} = \frac{\pi}{p_0}$$

Writing:

$$k = \frac{\xi \pi}{p_0} - \frac{\exp[(l - s_i)/\xi]}{1 + \exp[2(l - s_i)/\xi]}$$

We get:

$$\mu = \frac{1 - \sqrt{1 - 4k^2}}{2k}$$

When
$$(l-s_i) \gg \xi$$
 $\mu = \frac{p_0}{\pi \xi} - \sqrt{\frac{p_0^2}{\pi^2 \xi^2} - 1}$

In this last case we find the final value corresponding to equilibrium. This result explains the curve (Fig. 6) registered by the photomultiplier tube; a fixed μ means a fixed value of the angle at the surface, except in the regions where $(l-s_i) \sim \xi$ (wall "passing" through the surface).

The calculation of the velocity of the *i*th wall is not so easy because it is no more so obvious that we can neglect the border effects (Fig. 12). We can consider three possibilities in integrating the interaction between the *i*th wall and the surface:

a) take
$$\mu = \frac{p_0}{\pi \xi} - \sqrt{\frac{p_0^2}{\pi^2 \xi^2} - 1}$$
,

over the whole domain; we get the following form for the velocity:

$$\begin{split} V_i &= 8\omega_0 \xi \left\{ \exp\left[(s_i - s_{i-1})/\xi \right] - \frac{\mu \xi}{l - s_i} \right. \\ &\times \left\{ 1 - \exp\left[(s_i - l)/\xi \right] \right\} - \xi \left(2 - \frac{\pi^2 \xi}{p_0} \right) \middle/ 8(l - s_i) \right\} \end{split}$$

The shape of the drop is for this calculation fitted by a parabolic form.

- b) Take the whole expression of μ , knowing however that when $l-s_i \sim \xi$, the treatment is no longer valid, and one obtains the wrong value. The integration was made by computational techniques.
- c) Assume that the surface is very flat and put in simply the expressions for the distance $(l-s_i)$.

The comparison of this interpretation with experiment can be summed up rather simply:

1) for a wall located between two other walls (or between the rubbed surface and a wall) the agreement is satisfactory (Table 1). The theoretical values are very sensitive to each parameter and a

TABLE 1

Are present two walls: s_i and s_0 Experimental Data: $l=20.4\pm0.3\mu\mathrm{m}$ $H=10,600$ Gauss (±300 Gauss)										
P1 - 0.0 ± 0.2 µm										
$s_0~\mu{ m m}$	8.1	9.7	12	13.4	14.5	15.7				
$s_i \mu m$	14.8	16.4	17.5	18.1	18.7	19.2				
$V_0 \mu \mathrm{m min^{-1}}$	3.2	3.9	3.7	2.5	2.3	2				
$V_{\cdot um min^{-1}}$	3.2	2.7	1.8	1.2	1.1	1.0				

Theoretical: $l = 20.6 \, \mu \text{m} \, H = 10,600 \, \text{Gauss} \, K_{22} / \Delta \chi = 3.08 \, \text{emu cgs}$ $\gamma_1 / \Delta \chi = 0.9510^7 \, \text{emu cgs} \, p_0 = 9.5 \, \mu \text{m}$

$s_0 \mu \mathrm{m}$	8.1	9.7	12.05	13.35	14.4	15.7
$s_i \mu m$	14.8	16.04	17.6	18.294	18.9	19.2
$V_0 \mu \mathrm{m min^{-1}}$	3.4	4.1	3.5	2.5	2.0	-25
V_i a μ m min $^{-1}$	-22	- 27	- 34	- 34		
V_i b	21	23	5.2	8.1		
V_i e	7	5.8	1.7	2.6		

small variation of the field value, for instance within the experimental accuracy, changes drastically the value and even the sign of the velocity. Satisfactory results are obtained for the ratios

$$\frac{K_{22}}{\Delta_{\chi}} \simeq 3.0(8) \text{ emu cgs}$$

and:

$$\frac{\gamma_1}{\varDelta\chi}=0.95\;10^7\;\mathrm{emu\;cgs}$$

These values are in good agreement with other determinations. (10,13,14)

2) the description of the movement of the *i*th wall is less satisfactory: the (a) solution gives too strong a repulsive term, (b) a too strongly attractive one, and (c) always between the other two, without being so bad, seems not very realistic.

Let us note that in the case of an infinite broad film, the walls would adopt quickly an equilibrium value, and remain in the sample even for $H > H_C$. The surface variation is quite necessary to ensure the evolution toward the nematic state: it gives a positive contribution when:

$$rac{\xi \pi^2}{p_0} < 2$$
 That is $H > H_C = rac{\pi^2}{2} \sqrt{rac{K_{22}}{\varDelta \chi}} rac{1}{p_0}$

so we find a kinetic argument to introduce the field transition already given by De Gennes⁽¹⁵⁾ and R. B. Meyer.⁽¹¹⁾

In conclusion the dynamics of the cholesteric \rightleftharpoons nematic transition can be described by the continuum theory. It gives a simple and rather quick way of determining three ratios defining the medium. It could be used as a systematic tool allowing a rapid investigation of new products.

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