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FREEDERICKSZ TRANSITION OF NEMATICS IN AN OBLIQUE MAGNETIC FIELD

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Résumé. — Un film nématique planaire présente une transition de Fredericksz dans un champ magnétique perpendiculaire à l'orientation non perturbée. Cette transition se produit à H_{c_1} (distorsion en éventail) en champ perpendiculaire aux plaques et à H_{c_2} (torsion) en champ parallèle. Si le champ magnétique est appliqué à un angle oblique par rapport aux couches tout en restant perpendiculaire au directeur le seuil variera continuellement entre H_{c_1} et H_{c_2} . Nous calculons la distorsion au-dessus du seuil et comparons nos résultats à des expériences sur MBBA.

Abstract. — A planar nematic layer will show a Fredericksz transition in a magnetic field perpendicular to the unperturbed alignment. This transition occurs at $H = H_{c_1}$ (splay) in a perpendicular field and at $H = H_{c_2}$ (twist) in a field parallel to the layer. If the magnetic field is applied at an oblique angle with respect to the layer, but perpendicular to the direction of alignment, the threshold field can be varied continuously between H_{c_1} and H_{c_2} . We calculate the deformation pattern above the threshold and compare our results with experiments on MBBA.

1. **Introduction.** — A nematic layer thickness oriented uniformly parallel to the limiting plates (director \mathbf{n} along y) will undergo a Fredericksz transition in a magnetic field H applied perpendicular to the alignment (Fig. 1a) [1]. This results from a balance between the stabilizing elastic contribution and the destabilizing magnetic one (the susceptibility is anisotropic $\chi_a = \chi_{\parallel} - \chi_{\perp} > 0$ where \parallel and \perp refer to the direction of \mathbf{n} with respect to H). If H is perpendicular to the layers (along z), the initial distorsion above the critical threshold H_c is a pure splay and involves only a tilt of the director characterized by the polar angle with the horizontal $\varphi(z)$. The critical field determined by the splay elastic constant k_{11} is

$$H_{c_1} = (\pi/L) (k_{11}/\chi_a)^{1/2}.$$

If H is parallel to the slab (along x), the deformation is a pure twist (elastic constant, k_{22}) given by the polar angle $\omega(z)$ in the xy plane. The critical field is $H_{c_2} = (\pi/L) (k_{22}/\chi_a)^{1/2}$.

Here we study the more general case where H is still perpendicular to the initial unperturbed direction of alignment, but at an oblique angle θ with z . The

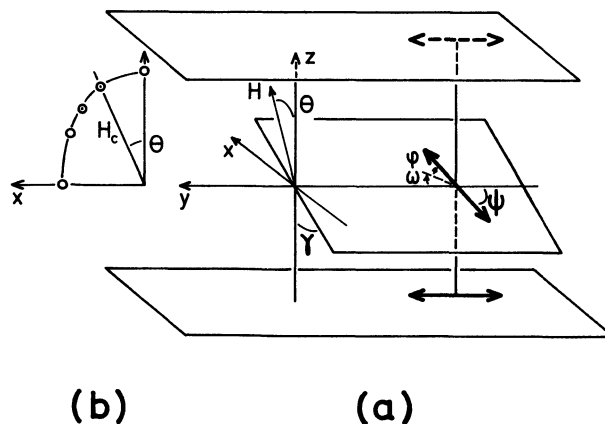


FIG. 1. — a) Geometry. The molecular orientation (\leftrightarrow) induced at the boundaries is distorted in the bulk (polar angles φ , ω). It takes place in a plane at an angle γ with z . H is perpendicular to y . b) Polar diagram of the $H_c(\theta)$ variation in the xz plane containing H .

second order phase transition taking place at the critical field is associated with the initial symmetry of \mathbf{n} with respect to H and, followingly, with the symmetric structure of the free energy with respect

to the angular variables : $G(\varphi) = G(-\varphi)$ (1.1) in the pure splay case, $G(\omega) = G(-\omega)$ (1.2) in the pure twist one.

In the general case discussed here, we will show that, for small distortions, the director remains parallel to a plane through the y axis and at an angle γ , a function of θ and of the elastic constants only, with the horizontal. Under these conditions, it is possible to describe completely the distortions by the angle $\psi(z)$ between \mathbf{n} and y . It is clear that $G(\psi) = G(-\psi)$. Thus, the existence of a second order transition with a critical field $H_c(\theta)$ follows from this symmetry. This would not be the case if H was not located in the xz plane. The case where H is in the yz plane has been studied [2, 3] and no transition with threshold is obtained.

The variation of the critical field $H_c(\theta)$, plotted on a polar diagram (Fig. 1b), is an ellipse with semi axis H_{c_1} (along z , $\theta = 0$) and H_{c_2} (along x , $\theta = \pi/2$). Experimental data points obtained on MBBA, a room temperature nematic used in this study, are also given.

In the following (section 2) we discuss a calculation of the threshold $H_c(\theta)$ and of the distortion for all values of fields $H(\theta)$ above threshold. The results extend those of a simple Landau like model valid for small distortions (appendix A). The results are used in section 3 for a comparison with experiments on nematic films, using the variation of the birefringence δ (connected to φ) and the rotation of the conoscopic image ε . A relation between the rotation ε and the angles $\varphi(z)$ and $\omega(z)$ is derived in appendix B.

2. Theory. — To calculate the deformation pattern above threshold we minimize G , the free energy per unit area of our sample. Let us denote the director by a unit vector $\mathbf{n}(z)$. The free energy per unit area is given by [1, 4]

$$G = \left(\frac{1}{2}\right) \int_0^L \{ k_{11}(\nabla \cdot \mathbf{n})^2 + k_{22}(\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + k_{33}(\mathbf{n} \times \nabla \times \mathbf{n})^2 - \chi_a(\mathbf{n} \cdot \mathbf{H})^2 \} dz. \quad (2.1)$$

The director can be expressed in terms of the angle of tilt $\varphi(z)$ and of twist $\omega(z)$ (see Fig. 1a)

$$\mathbf{n} = (\cos \varphi \sin \omega, \cos \varphi \cos \omega, \sin \varphi). \quad (2.2)$$

For the free energy G we obtain the expression

$$G = \left(\frac{1}{2}\right) \int_0^L \{ (k_{11} \cos^2 \varphi + k_{33} \sin^2 \varphi)(d\varphi/dz)^2 + (k_{22} \cos^2 \varphi + k_{33} \sin^2 \varphi) \cos^2 \varphi (d\omega/dz)^2 - \chi_a H^2 (\sin \theta \cos \varphi \sin \omega + \cos \theta \sin \varphi)^2 \} dz. \quad (2.3)$$

Minimizing the free energy, we obtain two non-linear differential equations of second order for the

functions $\varphi(z)$ and $\omega(z)$. We write these equations in a dimensionless form, introducing $\xi = z/L$ and the ratios of the elastic constants

$$\kappa = (k_{33} - k_{11})/k_{11}, \quad \alpha = (k_{33} - k_{22})/k_{22}, \quad \beta = (k_{11} - k_{22})/k_{22}. \quad (2.4)$$

These three parameters are not independent and

$$\beta = (\alpha - \kappa)/(1 + \kappa).$$

$$\begin{aligned} \frac{d}{d\xi} \left[(1 + \kappa \sin^2 \varphi) \frac{d\varphi}{d\xi} \right] &= \kappa \sin \varphi \cos \varphi (d\varphi/d\xi)^2 + \\ &+ [\alpha \sin \varphi \cos^3 \varphi - \sin \varphi \cos \varphi (1 + \alpha \sin^2 \varphi)] \\ &\times (d\omega/d\xi)^2 / (1 + \beta) - \pi^2 (H/H_{c_1})^2 \\ &\times (\sin \theta \cos \varphi \sin \omega + \cos \theta \sin \varphi) \\ &\times (\cos \theta \cos \varphi - \sin \theta \sin \omega \sin \varphi) \end{aligned} \quad (2.5)$$

$$\begin{aligned} \frac{d}{d\xi} \left[(1 + \alpha \sin^2 \varphi) \cos^2 \varphi \frac{d\omega}{d\xi} \right] &= \\ &= -\pi^2 (1 + \beta) (H/H_{c_1})^2 \times \\ &\times (\sin \theta \cos \varphi \sin \omega + \cos \theta \sin \varphi) \\ &\times \sin \theta \cos \varphi \cos \omega. \end{aligned} \quad (2.6)$$

To obtain the threshold field $H_c(\theta)$ we expand these two equations for small deformations, i.e. φ and $\omega \ll \pi/2$.

$$d^2\varphi/d\xi^2 = -\pi^2 (H/H_{c_1})^2 \cos \theta (\omega \sin \theta + \varphi \cos \theta) \quad (2.7)$$

$$d^2\omega/d\xi^2 = -\pi^2 (H/H_{c_1})^2 (1 + \beta) \times \sin \theta (\omega \sin \theta + \varphi \cos \theta). \quad (2.8)$$

For small deformations we can show that the director \mathbf{n} stays in a plane through the y -axis. Let this plane form an angle γ with the z -axis and let $\psi(\xi)$ be the angle of rotation of \mathbf{n} in this plane (see Fig. 1a). This gives us an additional relationship for $\varphi(\xi)$ and $\omega(\xi)$

$$\varphi(\xi) = \psi \cos \gamma; \quad \omega(\xi) = \psi \sin \gamma. \quad (2.9)$$

Inserting into eq. (2.7), (2.8) gives

$$\text{tg } \gamma = (1 + \beta) \text{tg } \theta \quad (2.10)$$

$$d^2\psi/d\xi^2 = -\pi^2 (H/H_{c_1})^2 (1 + \beta \sin^2 \theta) \psi. \quad (2.11)$$

With the boundary conditions $\psi(0) = 0$ and $d\psi/d\xi = 0$ at $\xi = 0.5$, we get the threshold field

$$H_c(\theta) = H_{c_1} (1 + \beta \sin^2 \theta)^{-1/2}. \quad (2.12)$$

This is indeed an ellipse with semi-axes H_{c_1} and $H_{c_2} = H_{c_1} \sqrt{1 + \beta}$. Writing the function $\psi(\xi)$ as

$$\psi(\xi) = \psi_m \sin(\pi \cdot \xi)$$

we can use (2.9) to obtain a Landau expansion of the free energy G in terms of the order parameter ψ_m .

In this way one can derive the threshold field (2.12) as well as the deformation pattern for H near $H_c(\theta)$. The details are given in appendix A.

To solve the Euler-Lagrange eq. (2.5), (2.6) numerically, we write them as four equations of first order introducing two functions $u(\xi)$ and $v(\xi)$. We then get

$$d\varphi/d\xi = v(\xi)/(1 + \kappa \sin^2 \varphi) \quad (2.13)$$

$$d\omega/d\xi = u(\xi)/(1 + \alpha \sin^2 \varphi) \cos^2 \varphi \quad (2.14)$$

$$du/d\xi = -\pi^2(1 + \beta)(H/H_{c1})^2 \times (\sin \theta \cos \varphi \sin \omega + \cos \theta \sin \varphi) \times \sin \theta \cos \varphi \cos \omega \quad (2.15)$$

$$dv/d\xi = \kappa \sin \varphi \cos \varphi (d\varphi/d\xi)^2 + [\alpha \sin \varphi \cos^3 \varphi - \sin \varphi \cos \varphi (1 + \alpha \sin^2 \varphi)] \times (d\omega/d\xi)^2 / (1 + \beta) - \pi^2(H/H_{c1})^2 \times (\sin \theta \cos \varphi \sin \omega + \cos \theta \sin \varphi) \times (\cos \theta \cos \varphi - \sin \theta \sin \omega \sin \varphi) \quad (2.16)$$

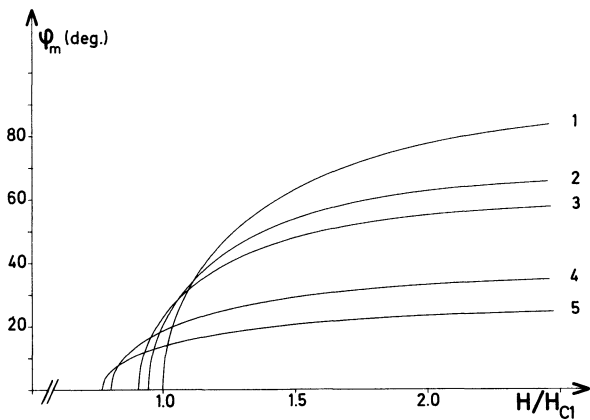


FIG. 2. — Calculated variation of the maximum tilt angle φ_m as a function of the reduced field H/H_{c1} where $H_{c1} = H_c(\theta = 0)$. Parameters $\kappa = 0.2$, $\beta = 0.86$, $\nu = 0.29$ obtained from experiments are used. In the figures 2, 3, 6, 7 the curves labelled 1, 2, 3, 4, 5, 6 correspond to $\theta = 0^\circ, 21.95^\circ, 30^\circ, 53.5^\circ, 60.25^\circ, 90^\circ$.

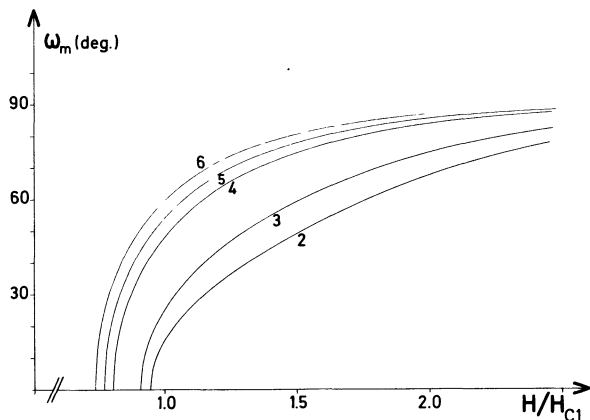


FIG. 3. — Calculated variation of the maximum twist angle ω_m versus H/H_{c1} .

The first two equations define the functions $v(\xi)$ and $u(\xi)$. Eq. (2.13), (2.16) can be solved by the Runge-Kutta method with the initial condition $\varphi = 0$, $\omega = 0$, $u = u_0$, $v = v_0$ at $\xi = 0$. The unknown parameters u_0 and v_0 are determined by the condition $d\varphi/d\xi = 0$ and $d\omega/d\xi = 0$ at $\xi = 0.5$.

Figures (2 and 3) show the results of such calculations. The maximum angle of tilt φ_m and the maximum angle of rotation ω_m are given as functions of H/H_{c1} for various values of θ . The values used for $\kappa = 0.2$ and $\beta = 0.86$ are those of MBBA as determined from experimental data presented below.

3. Experiments. — We have used high purity MBBA (methoxy *pn* benzilidene butyl anilin) which is nematic between 16° and 47°C . The experiments reported here were done at $25 \pm 1^\circ \text{C}$. The planar alignment was obtained by first evaporating under oblique incidence a thin SiO film in the inner sides of the glass limiting plates [5]. The oblique field is produced by a pair of Helmholtz coils giving the horizontal component of H , H_h and a solenoid of vertical axis giving the vertical one H_v (Fig. 5). The

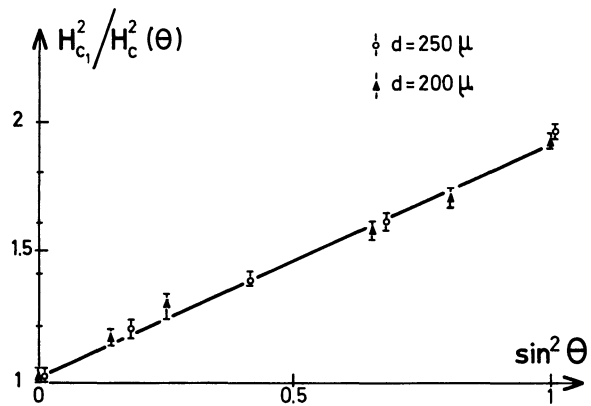


FIG. 4. — Variation of the critical field $H_c(\theta)$ in units giving a linear law as obtained from eq. (2.12). The limiting values are H_{c1} , ($\theta = 0$) and H_{c2} , ($\theta = \pi/2$).

angle θ is given by $\text{tg } \theta = H_h/H_v$. The distortion is followed by evaluating the changes of the characteristic conoscopic image from the double set of hyperbolas, centered at the vertical V of the laser beam, obtained in zero field :

The tilt angle $\varphi(z)$ is measured by counting the total number of fringes, δ , passing through point V as the field is increased from 0 to H .

$$\delta \cdot \lambda / n_e \cdot L = 1 - \int_0^{1/2} (1 + \nu \sin^2 \varphi)^{-1/2} d\xi \quad (3.1)$$

λ is the wavelength of light, L the sample thickness. The parameter giving the anisotropy of the refractive indices $\nu = (n_e/n_o)^2 - 1$. n_e and n_o are the extraordinary and ordinary index. For MBBA at 25° , $\nu = 0.29$ [6].

The angle of twist $\omega(z)$ is connected to the angle ε of rotation of the conoscopic image at point V. The result of the calculation given in appendix B shows that ε depends also on the splay plus bend distortion

$$\text{tg } 2\varepsilon = \frac{\langle \sin 2\omega(1 + \cos 2\varphi) \rangle}{\langle \cos 2\omega(1 + \cos 2\varphi) \rangle}. \quad (3.2)$$

The brackets mean an average over the cell thickness. The expression reduces to that calculated by de Gennes [7] in the case of pure twist.

Note : The determination of the critical field H_c and of the small distortion close to H_c involve long time constants [8]. Typically for $L = 200 \mu$ and $H/H_c = 1.1$, the exponential relaxation time constant is of the order of 500 s. Long enough times were used in order to get the static data points.

3.1 CRITICAL FIELDS. — The experimental results are given for two samples $L = 200, 250 \mu$ in figure (4) in reduced units $[H_c^2(\theta = 0)/H_c^2(\theta)]$ versus $\sin^2 \theta$. A linear variation, corresponding to the ellipse of figure (1b), is obtained in agreement with formula (2.12) which writes :

$$\begin{aligned} H_c^2(\theta = 0)/H_c^2(\theta) &= 1 + \sin^2 \theta \times \\ &\times [H_c^2(\theta = 0) - H_c^2(\theta = \pi/2)]/H_c^2(\theta = \pi/2). \end{aligned}$$

From the best linear fit of the slope we get $H_{c1}/H_{c2} = 1.36$, $k_{11}/k_{22} = 1.86$ and $\beta = 0.86$. The ratios agree well with published values [4].

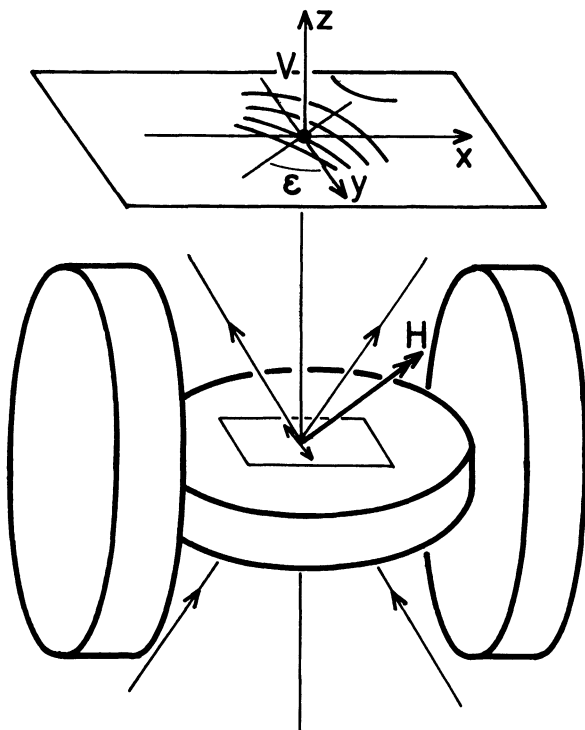


FIG. 5. — Experimental set up. Horizontal and vertical coils give the oblique field H perpendicular to the unperturbed molecular alignment along y . The projection in the upper plane of the conoscopic image from a monochromatic light converging on the sample allows the determination of both the integrated twist and birefringence.

3.2 DISTORTION. — Figures (6 and 7) give the experimental variation of δ and ε with H for different angles θ . The corresponding theoretical curves (solid lines) were obtained as follows : the parameter β is taken from the critical field variation. The parameter κ is obtained by a least square fit of the pure splay curve ($\theta = 0^\circ$). For the values of the parameters β , κ , ν , the differential equations (2.7, 2.8) were solved and δ and ε were calculated by numerical integration of (3.1, 3.2). The agreement of δ is good. The large spread of experimental data for the rotation ε comes from the visual estimate of this angle. The distorted conoscopic image is rather complex and does not have a symmetry axis (see Appendix B). This limits the accuracy on the evaluation of ε at point V.

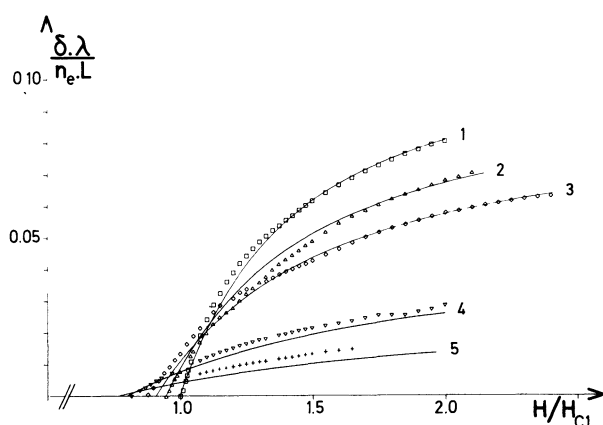


FIG. 6. — Experimental and calculated curves of the birefringence δ for a $L = 200 \mu$ thick sample. The normalized units are such that the results are independent of L . The parameters used in the calculations were obtained from the value of the ratio H_{c1}/H_{c2} and the best fit of the pure splay curve (1). The curves correspond to different θ values (see caption of Fig. 2).

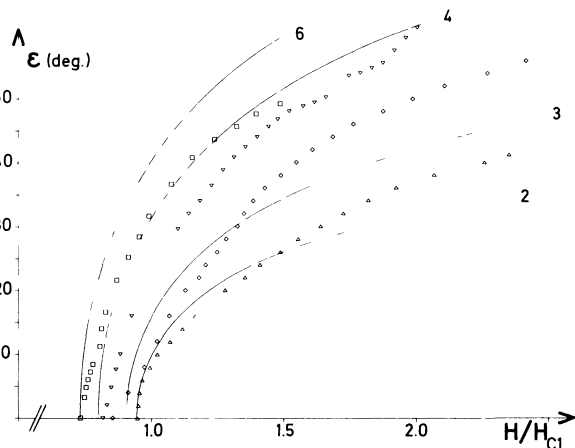


FIG. 7. — Experimental and calculated curves for the rotation of the conoscopic image ε and for different θ values.

The initial slope of the field variation of δ (Fig. 6) agrees with the direct calculation using the Landau expression. The asymptotic high field value can be obtained directly by expressing that the director is parallel to H across the sample.

In conclusion, in this oblique field study we have extended the range of the Freedericksz problem and have shown that the second order transition is retained although two variables appear in the description. Furthermore we can get the three Frank elastic constants k_{11}, k_{22}, k_{33} in a single experiment.

APPENDIX A. — Let us assume that for small deformations the director \mathbf{n} remains in a plane through the y -axis. This plane is at an angle γ with respect to the z -axis. Let $\psi(z)$ be the angle of rotation of \mathbf{n} in this plane. In terms of γ and $\psi(z)$, the director is

$$\mathbf{n} = (\sin \gamma \sin \psi, \cos \psi, \cos \gamma \sin \psi). \quad (\text{A. 1})$$

For small distortions $\psi(z) = \psi_m \sin \pi z/L$ and one gets a Landau like expansion

$$G = \pi^2/4 \{ (k_{11} \cos^2 \gamma)/L + (k_{22} \sin^2 \gamma)/L - [\chi_a LH^2 \cos^2 (\theta - \gamma)]/4 \} \psi_m^2 + \pi^2/16 \{ [(k_{33} - k_{11}) \cos^2 \gamma]/L + [\chi_a LH^2 \cos^2 (\theta - \gamma)]/16 \} \psi_m^4. \quad (\text{A. 2})$$

For a given value of γ one gets the critical field

$$H_c^2(\gamma) = \pi^2/\chi_a L^2 \cdot (k_{11} \cos^2 \gamma + k_{22} \sin^2 \gamma)/\cos^2 (\theta - \gamma)$$

and for $H \gtrsim H_c$

$$\psi_m^2 = [2 \cos^2 (\theta - \gamma) \chi_a (H^2 - H_c^2(\gamma))]/\sqrt{[(\pi^2/L^2) (k_{33} - k_{11}) \cos^2 \gamma + \chi_a H^2 \cos^2 (\theta - \gamma)]}.$$

In order to minimize G with respect to γ , we set $\partial G/\partial \gamma = 0$ which leads to

$$\text{tg } \gamma = (1 + \beta) \text{tg } \theta \quad (\text{A. 3})$$

$$H_c(\theta) = H_{c1} (1 + \beta \sin^2 \theta)^{-1/2} \quad (\text{A. 4})$$

$$\psi_m^2 = 4[H/H_c(\theta) - 1]/[1 + \kappa \cos^2 \theta/(1 + \beta \sin^2 \theta)]. \quad (\text{A. 5})$$

For the shift of birefringence one gets :

$$\delta \cdot \lambda/n_e \cdot L = v[(H/H_c(\theta)) - 1]/\sqrt{[1 + \kappa \cos^2 \theta/(1 + \beta \sin^2 \theta)]}. \quad (\text{A. 6})$$

The angle of rotation ε of the conoscopic image can be obtained in a similar way.

APPENDIX B. — We apply the adiabatic approximation valid when the distortion takes place over a large distance in front of the wavelength and expresses the ordinary (o) and extraordinary (e) waves as

$$e^{-i\Omega t} e^{i(k_x x + k_y y + k_z z)}.$$

In a layer dz we use the local axis ξ, η perpendicular to z such that the director is in the ηz plane

$$k_\xi = k_x \sin \omega(z) + k_y \cos \omega(z)$$

$$k_\eta = k_x \cos \omega(z) - k_y \sin \omega(z).$$

The surface of indices of the uniaxial material is composed of a sphere of radius n_o and of an ellipsoid of revolution around \mathbf{n} , of length axis n_o and n_e . We calculate the coordinates of a point of this surface, in the local reference frame. We write :

$$n_o = n(1 - u)$$

$$n_e = n(1 + u)$$

with

$$\frac{n_e + n_o}{2} = n, \quad \frac{n_e - n_o}{2n} = u$$

and consider only first order contribution in u ($< n/10$).

For the ordinary ray, we get :

$$k_{oz}^2 = \left(\frac{\Omega n_o}{c}\right)^2 - k_\eta^2 - k_\xi^2$$

and

$$k_{oz} \simeq \frac{\Omega n}{c} (1 - u) \times \left[1 - (k_\eta^2 + k_\xi^2) \frac{c^2}{2 \Omega^2 n^2} (1 + 2u) \right].$$

For the extraordinary ray, we have :

$$k_{ez} \simeq -\frac{\Omega}{c} u \xi \sin 2\varphi + \frac{\Omega n}{c} (1 + u \cos 2\varphi) \times \left[1 - \frac{c^2}{2 \Omega^2 n^2} (k_\eta^2 (1 - 2u) + k_\xi^2 (1 + 2u \cos 2\varphi)) \right].$$

For the layer dz , the phase shift is

$$d\rho = dz(k_{ez} - k_{oz}) = dz[n\Omega u(1 + \cos 2\varphi)/c + f k_\xi + a k_\eta^2 + b k_\xi^2]$$

with

$$a = c(3u - u \cos 2\varphi)/2 \Omega n$$

$$b = c(u - 3u \cos 2\varphi)/2 \Omega n$$

$$f = -u \sin 2\varphi.$$

We come back now to the reference axis x, y and we integrate over the thickness of the liquid crystal film

$$\rho(k_x, k_y) = d[n\Omega u \langle 1 + \cos 2\varphi \rangle / c + k_x \langle f \sin \omega \rangle + k_y \langle f \cos \omega \rangle + k_x^2 \langle b \sin^2 \omega + a \cos^2 \omega \rangle + k_y^2 \langle b \cos^2 \omega + a \sin^2 \omega \rangle]$$

where

$$\langle \quad \rangle = \frac{1}{d} \int_{-\frac{d}{2}}^{\frac{d}{2}} dz(\quad).$$

The first term gives the number of fringes

$$\delta = \frac{\rho - \rho(\varphi = 0)}{2\pi} = \frac{\Omega(n_e - n_o) d \langle 1 - \cos 2\varphi \rangle}{4\pi c}$$

The terms linear in k give the displacement of the center of the conoscopic image.

To evaluate the rotation ε of the conoscopic image we consider new cartesian axis X, Y

$$k_x = k_x \cos \varepsilon + k_y \sin \varepsilon$$

$$k_y = -k_x \sin \varepsilon + k_y \cos \varepsilon$$

such that the crossed contribution $k_x k_y$ cancels.

This gives

$$\begin{aligned} 2 \sin \varepsilon \cos \varepsilon \langle b \sin^2 \omega + a \cos^2 \omega \rangle - \\ - \langle b \cos^2 \omega + a \sin^2 \omega \rangle \\ + 2(\cos^2 \omega - \sin^2 \omega) \langle (b - a) \sin \omega \cos \omega \rangle = 0 \end{aligned}$$

or

$$\begin{aligned} \tan 2\varepsilon &= \frac{\langle (b - a) \sin 2\omega \rangle}{\langle (b - a) \cos 2\omega \rangle} \\ &= \frac{\langle (1 + \cos 2\varphi) \sin 2\omega \rangle}{\langle (1 + \cos 2\varphi) \cos 2\omega \rangle} \end{aligned}$$

Note that if we consider the perpendicular to the tangent of the curve passing through the origin ($k_x = k_y = 0$) it is given by

$$\tan \varepsilon' = \frac{\langle \sin 2\varphi \sin \omega \rangle}{\langle \sin 2\varphi \cos \omega \rangle}$$

This determination ε' reduces to ε to the limit of a planar or homeotropic sample, but is different in general. Numerical estimates of ε' following the method used in the article for ε , lead to values typically larger by 25% than ε . This ambiguity implies difficult measurements and explains the rather poor quantitative agreement on figure (7). Note that there is no such difficulty in the measurement of the birefringence.

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