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ANALYSIS OF THE OPTICAL REFLECTION ON AN INHOMOGENEOUS AND ANISOTROPIC
MEDIUM: APPLICATION TO A POISEUILLE FLOW IN A NEMATIC LIQUID CRYSTAL.

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Abstract

Some computations about the reflectivity of a light wave incident to an anisotropic and inhomogeneous medium are reported here. To explain experimental results concerning a nematic liquid crystal distorted by a Poiseuille flow, we introduce an absorption coefficient ($\chi = 10^{-4}$), simulating the global losses in the propagation of the electromagnetic plane wave. Maximal tilt angle and surface tilt angle are simultaneously revealed by two "critical" angles.

I. Introduction

In a previous article⁽¹⁾, we had shown the possibility of determining anchoring energies in a nematic liquid crystal from critical angle measurements. We had therefore supposed the optical homogeneity of the medium, which happens when the tilt angle variation within the layer is slow enough over a thickness equal to the penetration depth of the evanescent wave in the nematic liquid crystal. In general, we must take the distortion of the director in the vicinity of the surface as well as in the bulk into account, and study the optical reflection on such a distorted medium. We report here some computations about the reflectivity for a light wave incident to an anisotropic and inhomogeneous medium, and experimental results concerning its application to a Poiseuille flow study in a nematic liquid crystal cell.

II. Theoretical analysis

We consider a monochromatic homogeneous electromagnetic plane wave falling onto a plane boundary between two media : the former is an isotropic and homogeneous medium and the latter an anisotropic and inhomogeneous one (a liquid crystal distorted by an external field for example). The exact and theoretical determination of the reflectivity is quite difficult and we solve the problem by studying the reflectivity on birefringent multilayers. It is obvious that the number of layers used in our computations depends essentially on the anisotropic gradient index in the liquid crystal. We consider here a uniaxial and anisotropic medium and suppose that the orientation of the optic axis remains in the incidence plane ; therefore, for a T.M. wave, only the extraordinary wave needs to be taken into account⁽²⁾.

The geometry of the liquid crystal is defined as follows (Fig. 1): birefringent multilayers (the total thickness is d) surrounded by two isotropic, homogeneous and semi-infinite media (index N). These layers may be absorbing and their principal refractive indices are complex quantities:

$$\tilde{n}_o = n_o - i\chi_o \quad \text{for the ordinary wave}$$

and

$$\tilde{n}_e = n_e - i\chi_e \quad \text{for the extraordinary one.}$$

The equations describing transmission through and reflection from birefringent multilayers come from the Maxwell equations and the coupled-finite sequence method, used by Holmes and Feucht⁽³⁾, and extended to

absorbing layers is convenient to solve the problem. The numerical calculations have been carried out on a 370/168 IBM computer.

The number of layers (NC) used in our computations depends on the geometry of the distortion in the liquid crystal cell, and particularly the gradient index in the inhomogeneous medium. To avoid difficulties due to phase changes between two successive media, asymptotic solutions for the reflectivity have been always looked for. Another problem comes from the intense light scattering due to thermal fluctuations of the molecule orientation in nematics. In order to simulate this phenomenon, we introduce a strong absorption coefficient, thus describing all the losses in the propagation of the EM waves in the liquid crystal: absorption scattering due to the director fluctuations, scattering due to the roughness of the surfaces, static defects in the sample. To determine the values of the imaginary part of the refractive indices (the parameters χ_o and χ_e), we have performed an experiment with no distortion (the case of an anisotropic and homogeneous medium) and compared experimental results and computations obtained for different values of χ ; the best agreement occurs for $\chi = 10^{-4}$, but this value is not critical (similar results are obtained for $\chi = 2 \cdot 10^{-4}$). Such a description is not physically rigorous but allows to explain the experimental results. The corresponding attenuation constant is 24cm^{-1} in agreement with other experiments concerning the losses of a nematic liquid-crystal optical waveguide⁽⁴⁾. A more complete and exact formulation would be to choose different values for χ_o and χ_e , taking into account different scattering geometries, because it was found in experiments performed on MBBA that the intensity scattered depended on the angles between the director and the electric field⁽⁵⁾⁽⁶⁾⁽⁷⁾.

Now we consider a nematic confined between two parallel plates suitably treated in order to orient the optic axis perpendicularly to the layer (homeotropic configuration). When a Poiseuille flow occurs within the cell, the director alignment is modified. To describe this deformation, we make several assumptions:

- i) the boundary conditions are imposed (strong anchoring forces)
- ii) the three elastic constants provided from the continuum theory

are equal: $K_{11}=K_{22}=K_{33}=K$ (8)

- iii) the director remains in a plane perpendicular to the layers
(i.e. the incidence plane previously defined and corresponding to the flow direction)

The competition between the elastic and viscous torques leads to an equilibrium state given by the equation:

$$(1) \quad K \frac{d^2 \theta(z)}{dz^2} + \alpha_2 \frac{dv_x(z)}{dz} = 0$$

where $\theta(z)$ is the tilt angle defined with respect to the Ox axis (Fig.2), α_2 one of the Leslie coefficient and $v_x(z)$ the velocity (we suppose weak deformations of the director: $\theta \ll 1$). If η_{\perp} is the shear viscosity corresponding to the geometry of the cell ($\eta_{\perp} = \frac{1}{2} [-\alpha_2 + \alpha_4 + \alpha_5]$), we obtain from the Navier-Stokes equations:

$$(2) \quad \begin{aligned} \nabla_z p &= \rho g \\ \nabla_x p &= \eta_{\perp} \Delta v_x \end{aligned}$$

Then we find for the velocity:

$$(3) \quad v_x(\tilde{z}) = \frac{(-\nabla_x p) d^2}{8\eta_{\perp}} (1 - \tilde{z}^2)$$

where d is the thickness of the cell and \tilde{z} defined by: $\tilde{z} = \frac{2z}{d}$.

From (1) and (3) we obtain:

$$(4) \quad \theta(\tilde{z}) = -\frac{3\sqrt{3}}{2} \theta_{\max} \tilde{z} (\tilde{z}^2 - 1)$$

where θ_{\max} represents the maximal tilt angle:

$$\theta_{\max} = - \frac{(-\nabla_x p) \alpha_2 d^3}{72\sqrt{3} \eta_L K}$$

On the other hand, we obtain from (2):

$$\nabla_x p = - \frac{\rho g \Delta h}{L}$$

where L is the length of the cell, ρ the specific gravity of the liquid crystal and Δh the pressure difference between the two ends of the cell.

Then we can write:

$$(5) \quad \theta_{\max} = - \frac{\rho g d^3 \alpha_2 \Delta h}{72\sqrt{3} \eta_L K L}$$

We note that for $\alpha_2 < 0$ and $\nabla_x p < 0$, $\theta_{\max} > 0$ and the geometry acquired by the molecules of the liquid crystal is given in Figure 2.

If we take $d = 100 \mu\text{m}$, $K \sim 10^{-6} \text{dynes}$, $\rho \sim 1 \text{g/cm}^3$, $L = 4 \text{cm}$, $\Delta h = 1 \text{mm H}_2\text{O}$ and $(\alpha_2/\eta_L) \sim 1$, we obtain:

$$\theta_{\max} \sim 11 \text{ degrees}$$

We think this model valid for weak deformations and we shall see that experimental results agree with it. For a more complete analysis of the problem, we must take anchoring forces into account and solve the general problem of a Poiseuille flow in the nematic cell.

III. Experimental data

The Poiseuille cell was formed by a glass prism and a plate, the index of which is $N = 1.9250$ for the wavelength $\lambda = 0.5145 \mu\text{m}$ (Schott glass - LaSFN18). To obtain a good homeotropic alignment, a surface treatment was achieved with an appropriate surfactant (lecithine solution in ethanol). Tungsten wire spacers provide a constant thickness over all the cell and limit the hydrodynamic flux (this thickness was controlled by interferometric measurements). Then the cell is sealed with epoxy. The upper plate contains two cavities which act as reservoirs of liquid crystal and serve to prevent the formation of a meniscus inside the cell (Fig.3). Pressure measurement and regulation are identical to those used by Janossy, Pieranski and Guyon in their experimental study of the instabilities in nematics⁽⁹⁾: the flow is induced by applying a pressure difference between two small tubes connected with the upper plate; the accuracy is about $0.25 \text{ mm H}_2\text{O}$ on the pressure difference Δh .

The liquid crystal used in our experiment is the 4-cyano-4'-n-hexyl-biphenyl (6CB) obtained from BDH, the mesophase range being between 14°C and 29°C . All our measurements have been made at the ambient temperature ($T \sim 20^\circ\text{C}$) and the corresponding refractive indices, obtained from critical angle measurements⁽²⁾, were:

$$n_o = 1.544 \pm 0.001 \quad \text{and} \quad n_e = 1.709 \pm 0.001.$$

We have verified that these values were constant during all the time of the experiment, what is realized when $\Delta T < 0.3^\circ\text{C}$ (for $T \sim 20^\circ\text{C}$, the thermal gradient index $\Delta n/\Delta T \sim 3.10^{-3}/^\circ\text{C}$).

The incident light comes from an argon laser used for the wavelength $\lambda = 0.5145 \mu\text{m}$. A half-wave plate can be used to change the polarisation

if necessary. To reduce the beam divergence, the laser waist is put on the level of the interface prism-liquid crystal by means of a single lens. The resulting beam divergence is less than $1'$ of arc. The experimental arrangement is very similar to that used in the determination of liquid crystal refractive indices, but photometric measurements were achieved with a PIN-10DP photodiode; the resulting absolute accuracy is 0.01 on the value of the reflectivity.

IV. Results and discussion

Experimental results are given in Figure 4 for different values of Δh ; we find that interference fringes are not visible in the undisturbed medium and this is due to the light scattering and the thickness of the cell. Now when a distortion occurs in the liquid crystal, due to the flow in the cell, the reflectivity R is strongly modulated versus the incidence angle i and fringes appear, the number and the angular spacing of which depend on Δh . Moreover, there exists an incidence angle i_m , corresponding to the complete disappearance of these fringes and easy to detect (visually or by means of a detector); the accuracy in the determination of i_m is about $1'$ of arc. To explain this phenomenon, we consider three cases (Fig. 5):

i) $i > i_l$: if the incidence angle is greater then the angle corresponding to the homeotropic configuration (defined by: $n_e = N \sin i_l$), the light is totally reflected by all the layers present in the liquid crystal.

ii) $i_l > i > i_m$: then the light is partially transmitted in the first layers but there exists several layers inside the cell on which the incident wave is totally reflected. The angle i_m corresponds to the maximal deformation of the optic axis θ_{\max} and is given by:

$$(6) \quad N^2 \sin^2 i_m = n_o^2 \sin^2 \theta_{\max} + n_e^2 \cos^2 \theta_{\max}$$

iii) $i < i_m$: the light is transmitted through all the layers and reaches the last interface defined by the upper plate. The fringes resulting from the interference phenomenon between the two plates are not very visible because of the light scattering in the nematic cell.

In our experiment, the total thickness was: $d = 97 \pm 1 \mu\text{m}$, and the maximal deformation θ_{max} occurred in a layer at a distance of $20.5 \mu\text{m}$ from the interface prism-liquid crystal. We note this distance doesn't depend on Δh .

In order to confirm our hypothesis, computations have been achieved by means of the previously described method. The results appear in Figure 6 for the following values:

$$n_o = 1.5442 - 0.0001 i$$

$$n_e = 1.7084 - 0.0001 i$$

$$N = 1.925$$

$$d = 97 \mu\text{m}$$

$$\theta_{\text{max}} = 10.3 \text{ degrees.}$$

The corresponding critical angles are:

$$i_l = 62^\circ 33'$$

$$i_m = 62^\circ 14' .$$

The computations agree quite well with our measurements but we must make a few remarks about them and discuss the influence of the different

parameters on the numerical results. A slight increase of θ_{\max} produces expansion of the fringes but doesn't affect their amplitudes; if the variation is stronger (when the pressure difference increases) fringes appear more numerous between i_l and i_m . Now if we modify one of the refractive indices, n_e , the corresponding effect is a global translation of the entire curve. A most delicate point remains the value of the parameter χ previously defined as the imaginary part of the refractive indices of the nematic liquid crystal. In the case of strong attenuations ($\chi \sim 10^{-2}$), the fringes are no more visible and for weak attenuations ($\chi \sim 10^{-5}$), the modulation increases. The limit $\chi = 0$ corresponding to an anisotropic and non-absorbing medium also produces the disappearance of the phenomenon because the total reflection is not attenuated, the gradient index being not sufficiently important to consider the medium as an inhomogeneous one. Then we can say that the fringes are revealed by the scattering of the light in the nematic cell, simulated by the introduction of the parameter χ in our computations.

We have shown it was possible to measure the maximal deformation of the liquid crystal (θ_{\max}) as a function of the pressure difference between the two extremities of the cell (Δp , or Δh), the results are given in Figure 7 and in the following table:

Δh (mm H_2O)	θ_{\max} (degrees)
0.5	7.1 ± 0.4
1.0	14.4 ± 0.3
1.5	19.3 ± 0.2
2.0	26.6 ± 0.2
2.5	33.9 ± 0.2
3.0	41.1 ± 0.2
3.5	47.8 ± 0.2

The value for $\Delta h = 1 \text{ mm H}_2\text{O}$ is different from the one previously given (10.3 degrees) because of the uncertainty on Δh and also because the temperature of the cell was different in the two experiments; for this latter reason, it would be interesting to investigate the temperature dependence of the tilt angle θ_{\max} .

When $\theta_{\max} > 10$ degrees, we cannot assume $\theta \ll 1$ and the previous model is no more valid: we must replace the equation (1) by the following one provided from the Eriksen-Leslie equations^(10,11):

$$\begin{aligned} (K_{11}\sin^2\theta + K_{33}\cos^2\theta) \frac{d^2\theta}{dz^2} + (K_{33}-K_{11})\sin\theta\cos\theta\left(\frac{d\theta}{dz}\right)^2 \\ + (\alpha_2\cos^2\theta - \alpha_3\sin^2\theta) \frac{dv_x}{dz} = 0 \end{aligned}$$

Moreover, the viscosity η used in the equation (2) is different from the Miesowicz viscosity η_{\perp} ^(12,13). The effective viscosity is intermediate between η_{\perp} and η_{\parallel} (where $\eta_{\parallel} = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6)$). However, it is possible to deduce an accurate value of the ratio α_2/η_{\perp} from these measurements (the initial slope of the curve giving Δh as a function of θ_{\max}):

$$\alpha_2/\eta_{\perp} = 1.23 \pm 0.07$$

Other experiments would be necessary to determine the maximal value of θ_{\max} which is the angle defined by $\tan^2\theta_{\max} = \alpha_2/\alpha_3$, and first determined by Ghhwiller on MBBA⁽¹⁴⁾. We could also study the influence of the anchoring forces on the deformation of the liquid crystal molecules, using higher differences of pressure, in order to analyse the director field in the boundary layers. For all these reasons, we think our method

provides a convenient and original tool to investigate hydrodynamics problems in nematics.

V. Conclusion

In this work, we have presented a new optical method to analyse the distortion of the liquid crystal in the case of a Poiseuille flow. The interesting point concerns the possibility to observe independently and simultaneously the surface behavior and the bulk behavior of the director, clearly demonstrated by the existence of the two critical angles: i_1 and i_m . We hope these results will stimulate further experiments for a better understanding of physical properties of liquid crystals and particularly their surface properties.

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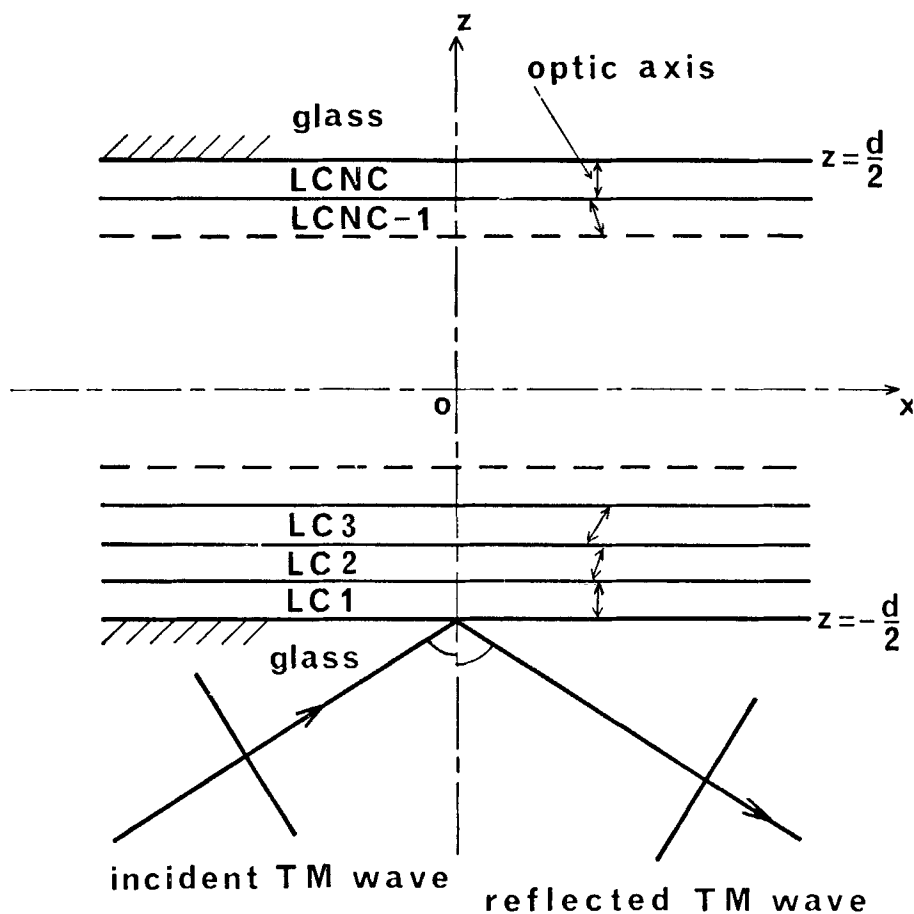


Figure 1: Schematic diagram showing the model used in the computations.

(we note NC as the number of the thin layers and d the thickness of the cell).

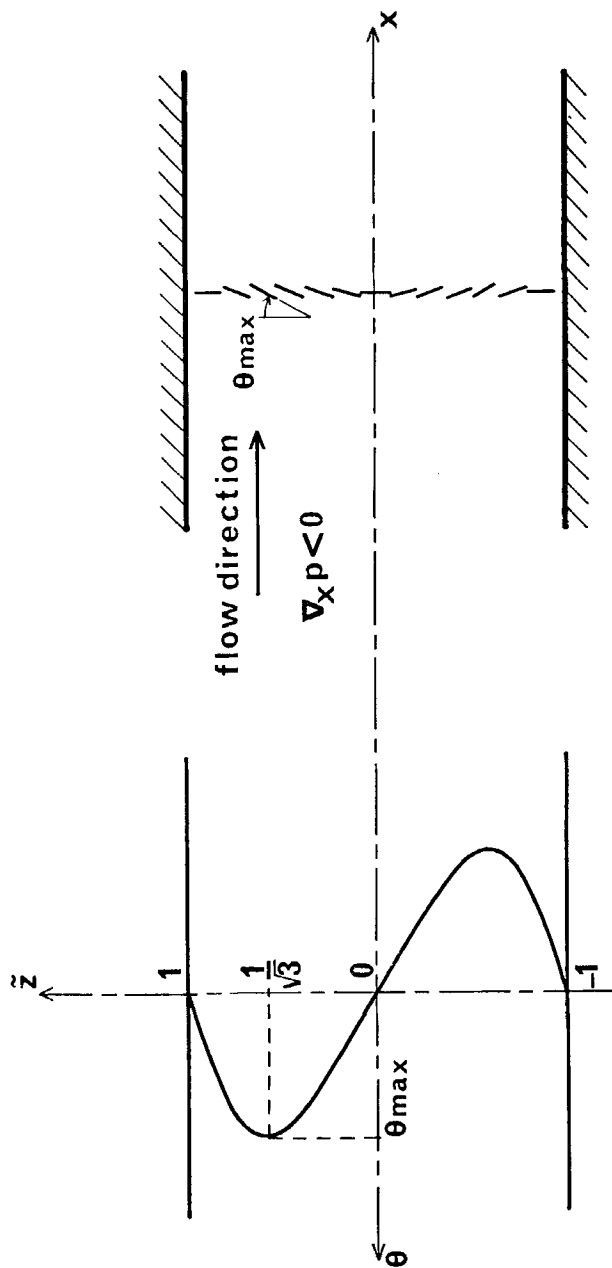


Figure 2: Tilt angle θ versus reduced coordinate \tilde{z} and corresponding representation of the liquid crystal molecules in the cell.

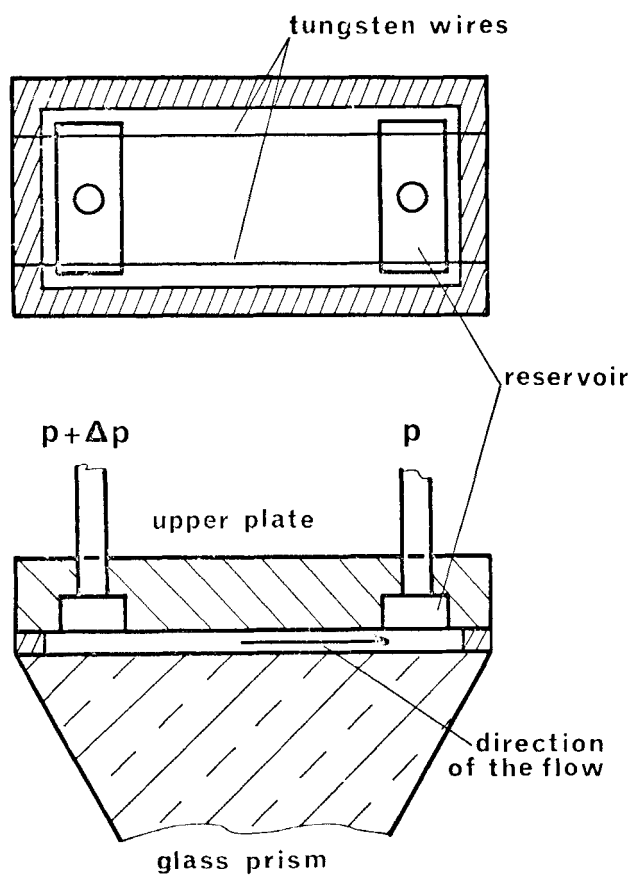


Figure 3: Experimental arrangement used for the Poiseuille cell.

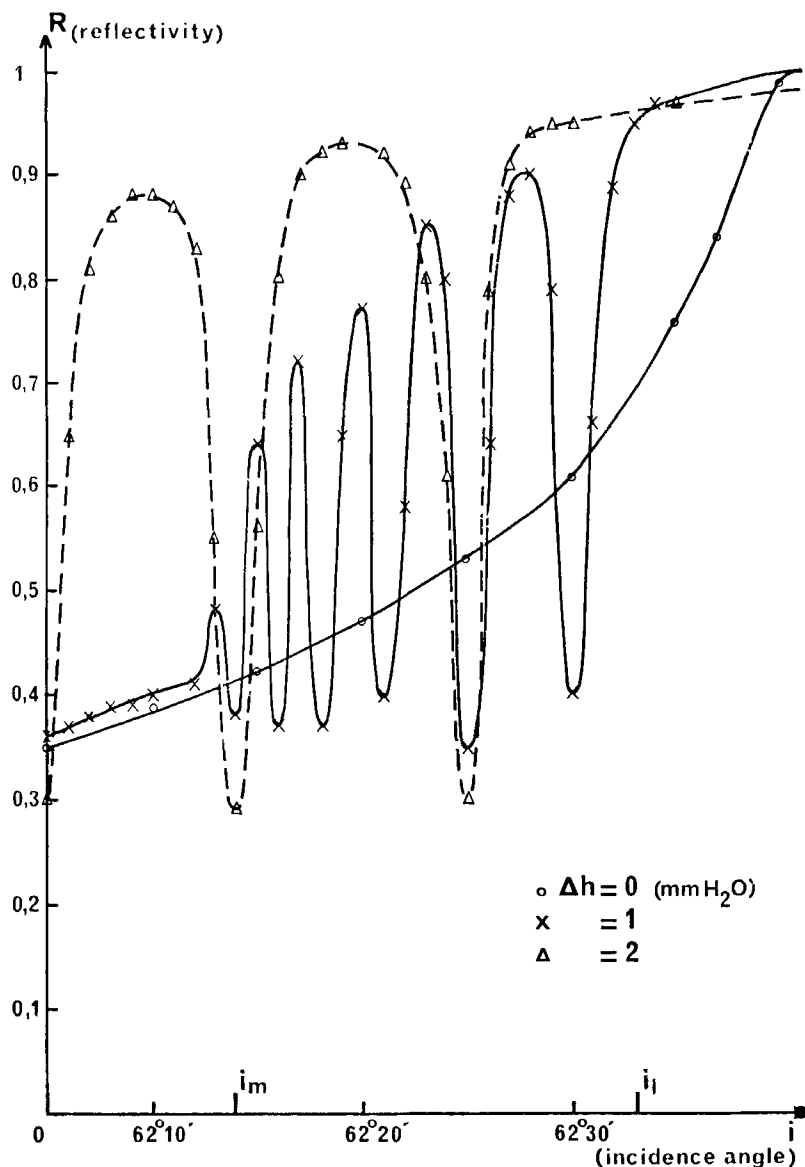


Figure 4: Experimental curves giving the reflectivity as a function of the incidence angle for different values of the pressure difference Δh .

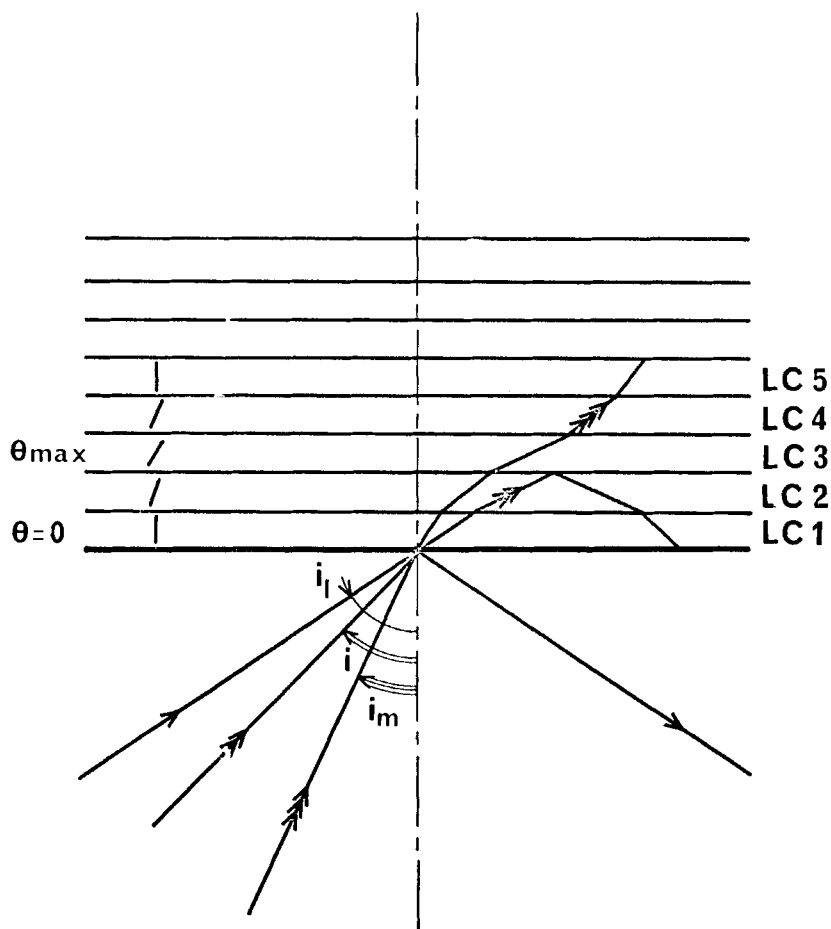


Figure 5: Diagram explaining the propagation of the wavelight through the layers for different incidence angles.

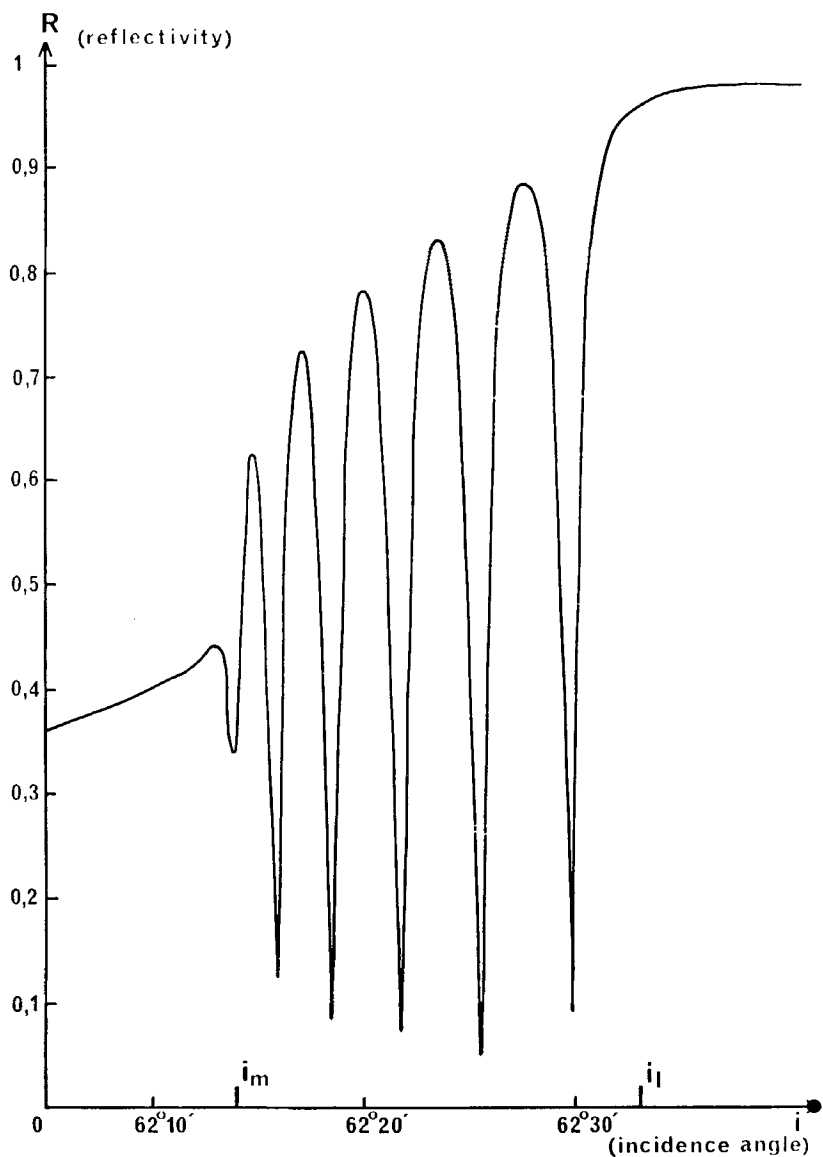


Figure 6: Theoretical curves giving the reflectivity as a function of the incidence angle for the following values: $\theta_{\max} = 10.3$ degrees; $d = 97 \text{ } \mu\text{m}$; $N = 1.925$; $\tilde{n}_o = 1.5442 - 0.0001i$; $\tilde{n}_e = 1.7084 - 0.0001i$

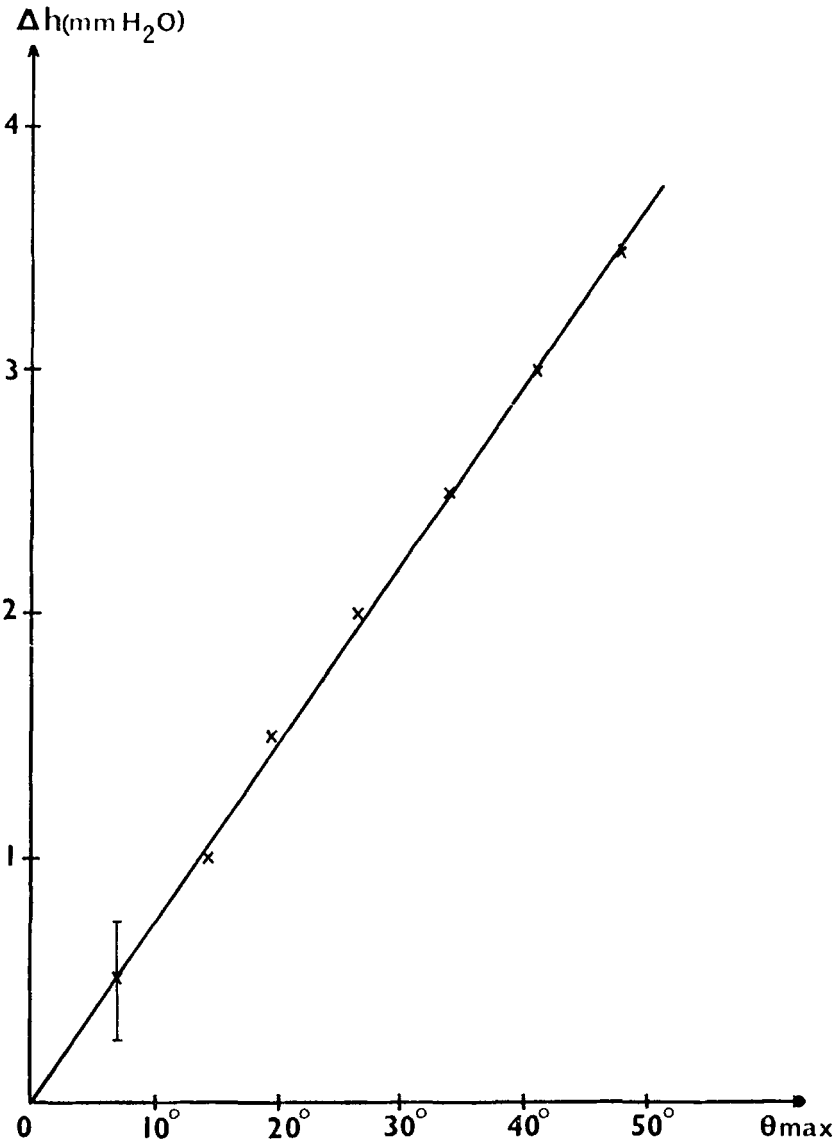


Figure 7: Curves giving (Δh pressure difference) as a function of θ_{max} (tilt maximum angle in the nematic cell).

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