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FREEDERICKSZ TRANSITION OF A HOMEOTROPIC NEMATIC LIQUID CRYSTAL IN ROTATING MAGNETIC FIELDS

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Résumé. — Nous présentons une étude théorique et expérimentale de la transition de Freedericksz d'une lamelle nématique homéotrope soumise à un champ magnétique tournant dans le plan de la lame, avec une vitesse angulaire ω . Deux champs seuils sont nécessaires pour décrire le comportement du directeur :

- Un champ seuil d'accrochage $H_a(\omega)$.
- Un champ seuil pour l'apparition de la distorsion.

Ils sont égaux pour une fréquence limite ω_1 .

Si $\omega < \omega_1$, $H_a(\omega) < H_c(\omega)$ et la distorsion apparaît dans un régime où le directeur tourne de façon synchrone avec le champ.

Si $\omega > \omega_1$, $H_a(\omega) > H_c(\omega)$, et on peut observer un régime synchrone et un régime asynchrone de rotation du directeur.

Abstract. — We present experimental and theoretical investigations on the Freedericksz transition of a homeotropic nematic slab in rotating magnetic field. Two characteristic fields, and one critical frequency are necessary to describe the behaviour of the sample :

- $H_a(\omega)$ separates synchronous and asynchronous rotations of the director.
- $H_c(\omega)$ is the threshold for the distortion.

$H_a(\omega) = H_c(\omega)$ for $\omega = \omega_1$.

For $\omega < \omega_1$, $H_a(\omega) < H_c(\omega)$, and only a synchronous regime is observed.

For $\omega > \omega_1$, $H_a(\omega) > H_c(\omega)$ and both synchronous and asynchronous regime appear.

1. Introduction. — If a magnetic field \mathbf{H} is applied perpendicular to the optical axis of a nematic slab, the average direction of orientation of the molecules is distorted for H larger than a threshold field H_{c_0} . For the homeotropic configuration (Fig. 1)

$$H_{c_0} = \frac{\pi}{d} \sqrt{\frac{K_3}{\chi_a}},$$

where d is the sample thickness, K_3 the Frank elastic constant for bend distortions and χ_a the anisotropic part of the diamagnetic susceptibility [1]. As the distortion appears, the director \mathbf{n} remains in the plane $(\mathbf{H}, \mathbf{n}_0)$, and the configuration is entirely defined by the angle $\theta(z)$ between \mathbf{n} and \mathbf{n}_0 (Fig. 1). $\theta(z)$ can be determined by writing the equilibrium between the elastic and the magnetic torques acting on the director.

The case of a rotating field is more complex. The Freedericksz transition in a rotating field has previously been studied by Gasparoux and Canet [2], but their

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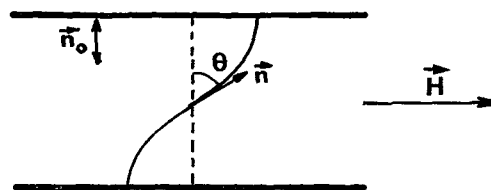


Fig. 1. — Distortion of the molecular alignment in a static Freedericksz transition. The director \mathbf{n} remains in the $(\mathbf{H}, \mathbf{n}_0)$ plane, and is characterized by one angle θ .

geometry is complex (planar anchoring of the molecules) and their results are not yet understood.

We have chosen the homeotropic geometry. In that case, the normal orientation of the molecules at the glass plates provides a pivot layer for the molecular rotation, and allows for a uniform distortion in the whole sample.

— If \mathbf{H} rotates very slowly, \mathbf{n} remains in the rotating plane $(\mathbf{H}, \mathbf{n}_0)$, and the distortion appears for $H = H_{c_0}$.

— If \mathbf{H} rotates at infinite frequency, we have something similar to the Freedericksz transition with an electrical field $\mathbf{E} = \mathbf{H}/\sqrt{2}$, normal to the plates, and a

negative dielectric anisotropy $\epsilon_a = -\chi_a$ (complete degeneracy for the tilt direction in the plane of the slab). The distortion appears for $H_{c\infty} = \sqrt{2} H_{c0}$.

— The present paper essentially shows experimental and theoretical investigations of what happens between those two limiting cases. We shall see that two thresholds are necessary to describe the behaviour of the director :

1) $H_a(\omega)$, threshold for the synchronous rotation of the molecules with the field.

2) $H_c(\omega)$, threshold for the Freedericksz distortion.

2. **General equation.** — In the static case, we have seen that one angle $\theta(z)$ is necessary to describe the distorted configuration. In the rotating case, \mathbf{n} is defined by two angles, θ and φ (Fig. 2). The motion

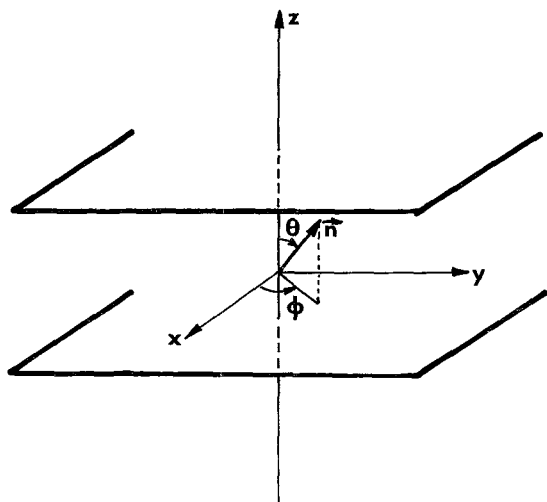


FIG. 2. — In the case of rotating field the plane of the distortion rotates, and is characterized by φ .

equations for θ and φ are given by the equilibrium between the Frank elastic torque, the Leslie viscous torque, and the magnetic torque acting upon the director [3] :

$$\begin{aligned} \gamma_1 \left\{ \frac{\partial \varphi}{\partial t} \sin \theta \begin{bmatrix} -\cos \theta \cos \varphi \\ -\cos \theta \sin \varphi \\ \sin \theta \end{bmatrix} + \frac{\partial \theta}{\partial t} \begin{bmatrix} -\sin \varphi \\ \cos \varphi \\ 0 \end{bmatrix} \right\} = \\ = \chi_a H^2 \sin \theta \cos(\omega t - \varphi) \begin{bmatrix} -\cos \theta \sin \omega t \\ \cos \theta \cos \omega t \\ \sin \theta \sin(\omega t - \varphi) \end{bmatrix} + \\ + f(\theta) \begin{bmatrix} -\sin \varphi \\ \cos \varphi \\ 0 \end{bmatrix} \end{aligned} \quad \begin{matrix} (1a) \\ (1b) \\ (1c) \end{matrix}$$

Here

$$\begin{aligned} f(\theta) = \frac{\partial^2 \theta}{\partial z^2} (K_1 \sin^2 \theta + K_3 \cos^2 \theta) \\ + \left(\frac{\partial \theta}{\partial z} \right)^2 (K_1 - K_3) \sin \theta \cos \theta, \end{aligned}$$

and γ_1 is an effective viscosity for the rotation of the director, taking backflow effects into account [4]. The velocity field is normal to the rotating plane (\mathbf{n}_0, \mathbf{n}) of the distortion and confined to two boundary layers of thickness of the order

$$\xi(H) = \frac{1}{H} \sqrt{\frac{K_3}{\chi_a}}$$

At high fields, these boundary layers are thin compared to d , and backflow corrections to γ_1 are negligible.

An important simplification comes from the fact that eq. (1c) gives the evolution of φ , independently of θ . Thus we shall first discuss the evolution of φ , supposing that the distortion exists, and then we shall discuss the evolution of θ .

3. **Threshold field for the synchronous rotation $H_a(\omega)$.**

— The eq. (1c) can be written as

$$\tau \dot{\varphi} = \sin 2(\omega t - \varphi) \quad (2)$$

where τ is a characteristic time of the nematic

$$\tau = \frac{2 \gamma_1}{\chi_a H^2},$$

$\omega \tau = 1$ defines a characteristic field

$$H_a(\omega) = \left(\frac{2 \gamma_1 \omega}{\chi_a} \right)^{1/2}.$$

3.1 $H > H_a(\omega)$ ($\leftrightarrow \omega \tau < 1$). — The nematic follows the rotation of the field with a constant retardation angle α (Fig. 3).

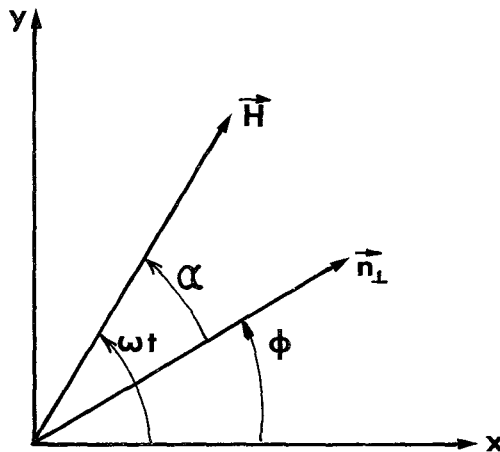


FIG. 3. — The plane of the distortion is at an angle α with the $(\mathbf{H}, \mathbf{n}_0)$ plane. In the synchronous regime, α is constant. In the asynchronous regime, α increases continuously with time, with mean angular velocity $2\pi/T$.

The solution of (2) is given by

$$\begin{cases} \varphi = \omega t - \alpha \\ \sin 2\alpha = \omega \tau. \end{cases}$$

We have a synchronous regime : the director rotates with the angular velocity of the field ω .

Experimentally, this regime is rather easy to characterize: with a polarizer at an angle α with respect to the field, and a crossed analyzer, a stable extinction of the transmitted intensity is obtained for a given field H_α at each frequency. The corresponding equal retardation angle curves are shown in the H^2 versus ω diagram of figure 4, for $\alpha_1 = 10^\circ$ and $\alpha_2 = 20^\circ$.

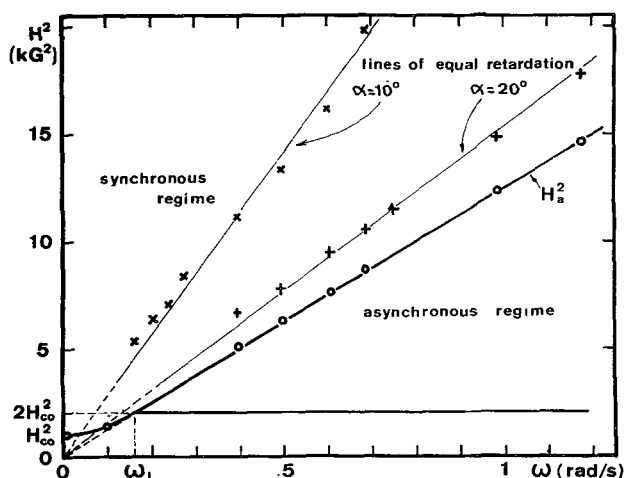


FIG. 4. — Phase diagram of an homeotropic nematic slab in a rotating field H : above the line $H_c(\omega)$, a distortion appears; the line $H_a(\omega)$ separates the synchronous and the asynchronous rotation of the molecules. In the synchronous regime, we represent the lines of constant α ($H_a(\omega)$ is the line of $\alpha = \pi/4$). In this H^2 versus ω diagram, they are straight lines as predicted theoretically.

They are observed to be straight lines. The line $\alpha = \pi/4$ defines $H_a(\omega)$, and limits the synchronous regime.

3.2 $H < H_a(\omega)$ ($\leftrightarrow \omega\tau > 1$). — Now the characteristic time τ for the rotation of the molecules is long compared to the periodicity of the field, and the molecules cannot follow the too rapid rotation of the field. The retardation angle is no longer constant in time.

The solution of eq. (2) is given by

$$\left\{ \begin{array}{l} \varphi = \omega t - \alpha(t) \\ \operatorname{tg} \alpha(t) = \frac{1}{\omega\tau} + \left(1 - \frac{1}{\omega^2\tau^2}\right)^{1/2} \times \\ \quad \times \tan\left(\frac{t}{\tau}(\omega^2\tau^2 - 1)^{1/2} + \varphi_0\right). \end{array} \right. \quad (3)$$

With a polarizer parallel to the field, and a crossed analyzer, we can observe $\sin^2(\alpha(t))$ as the intensity of transmitted light.

$\sin^2(\alpha(t))$ is a periodic function of time, with a periodicity

$$T = \frac{\pi\tau}{(\omega^2\tau^2 - 1)^{1/2}}. \quad (4)$$

This gives a convenient linear relationship between T^{-2} and H^4 :

$$\frac{\pi^2}{\omega^2 T^2} = 1 - \frac{H^4}{H_a^4(\omega)}. \quad (5)$$

Figure 5 shows a typical recording of the transmitted intensity through the sample, in the case where we have a distorted configuration, but H smaller than $H_a(\omega)$.

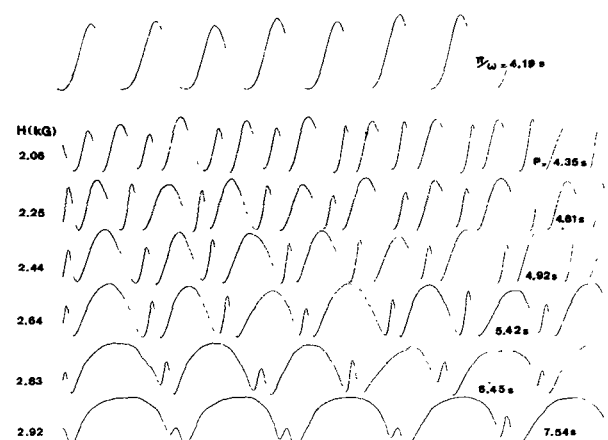


FIG. 5. — Typical recording of the transmitted intensity with a polarizer parallel to H , and a crossed analyzer, in the asynchronous regime. It is a periodic function of the time. The period T diverges as H goes to $H_a(\omega)$.

The exact form of the curve depends on both α and birefringence effects (depending on the time variation of θ) which changes the color of the transmitted light. However, the period T and the two semi-periods

$$\left(0 < \alpha < \frac{\pi}{2}, \quad \text{and} \quad \frac{\pi}{2} < \alpha < \pi\right)$$

are well defined.

Figure 6 shows a plot of $\frac{\pi^2}{\omega^2 T^2}$ versus H^4 , for different values of ω . The linear behaviour predicted by

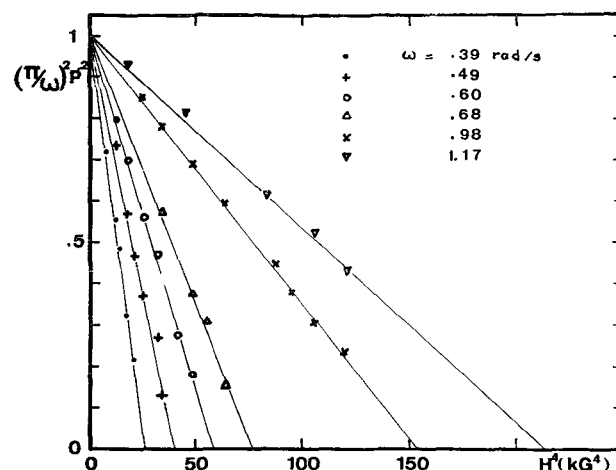


FIG. 6. — The linear dependence of $1/T^2$ versus H^4 gives a determination of the threshold for synchronous rotation $H_a(\omega)$.

eq. (5) is well observed, and allows for an accurate determination of $H_a(\omega)$ (Fig. 4) (*).

The field dependence of the relative values of the semi-periods is also in agreement with eq. (3).

4. Threshold field for the distortion $H_c(\omega)$. — The evolution of the Freedericksz distortion is given by eq. (1a, b), and can be written

$$\gamma_1 \frac{\partial \theta}{\partial t} = \chi_a H^2 \cos^2 \alpha \sin \theta \cos \theta + f(\theta). \quad (6)$$

Two kinds of behaviour are expected, depending on whether α is constant or not :

4.1 SYNCHRONOUS REGIME $H_c(\omega) > H_a(\omega)$. — If the distortion appears in the synchronous rotation regime, the retardation angle α is constant, and we have something equivalent to the static case, with an effective field $H \cos \alpha$, smaller than H .

The threshold field for the distortion is then given by $H_c(\omega) = H_{c_0}/\cos \alpha$, i. e.

$$\frac{H_c^2(\omega)}{H_{c_0}^2} = 1 + \frac{\omega^2}{\omega_1^2}$$

with $\omega_1 = \frac{K_3 \pi^2}{\gamma_1 d}$, representing the limiting frequency

for this synchronous regime at threshold. Experimental determination of $H_c(\omega)$ in the synchronous regime is shown on the left part of figure 4. For $\omega = \omega_1$, $H_c(\omega) = H_a(\omega)$, and the distortion appears at $H_c(\omega_1)$ with the maximum retardation angle $\alpha = \pi/4$.

4.2 ASYNCHRONOUS REGIME $H_c(\omega) < H_a(\omega)$. — For $\omega > \omega_1$, the distortion appears in the asynchronous regime. α is modulated in time, and eq. (6) becomes much more complex. In order to discuss the appearance of the distortion, we look at the evolution of the thermal fluctuation of wave vector $q_{\perp} = 0$, $q_z = \pi/d$ which is first destabilized at the Freedericksz transition.

With $\theta = \theta_M \cos \pi z/d$, eq. (6) can be written

$$\left(\cos^2 \alpha(t) - \xi^2 \frac{\pi^2}{d^2} \right) \theta_M = \frac{\tau}{2} \frac{d\theta_M}{dt}$$

where ξ is the magnetic coherence length $\xi = \frac{1}{H} \sqrt{\frac{K_3}{\chi_a}}$.

The fluctuation becomes unstable for

$$S = \frac{2}{\tau} \left(\cos^2 \alpha - \frac{\xi^2 \pi^2}{d^2} \right) > 0$$

S is modulated in time, with the periodicity T .

— For $H < H_{c_0}$, S is always negative, and the nematic remains immobile.

(*) Another way to measure τ is to rotate the sample quickly by a small angle α_0 relative to the field (now static), and watch the relaxation of the director back to $\alpha = 0$. The transmitted light intensity now decays exponentially to zero with a relaxation time $\tau/4$.

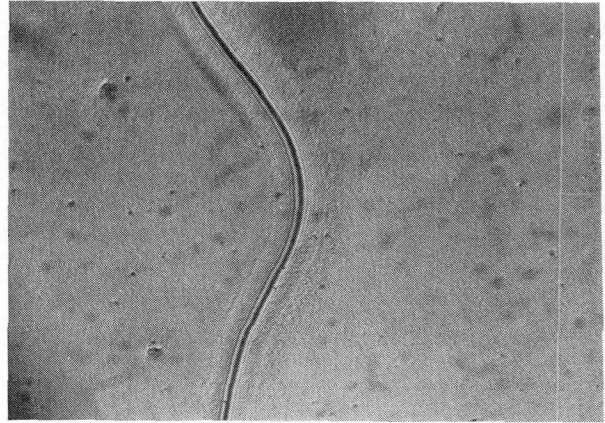
— It can be shown that the Freedericksz distortion can occur only if $\langle S \rangle > 0$ [5].

This defines the threshold for the distortion :

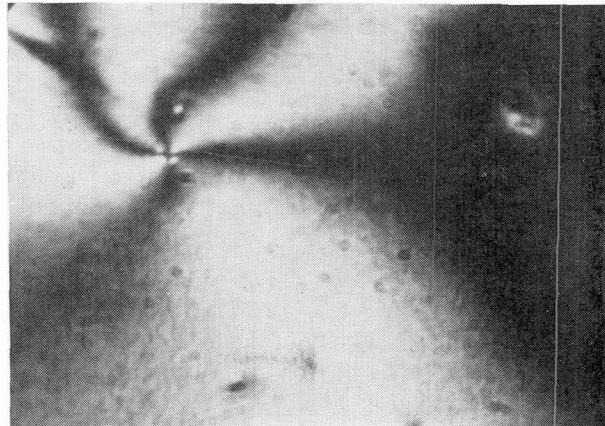
$$H_c(\omega) = \sqrt{2} H_{c_0}.$$

For $H > \sqrt{2} H_{c_0}$, the distortion has a finite amplitude.

(a)



(b)



(c)

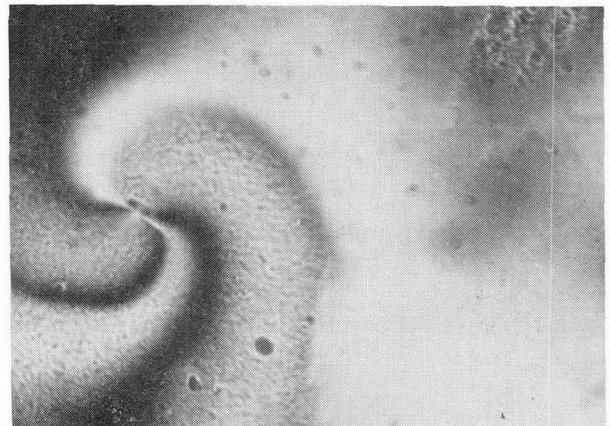


FIG. 7. — Defects observed at the Freedericksz transition. The degeneracy for the distortion depends on the frequency : a) at $\omega = 0$, walls ; b) at ω^∞ , umbilics ; c) at intermediate ω , spirals.

For $H_{c_0} < H < \sqrt{2} H_{c_0}$ there is no finite distortion but the thermal fluctuations are amplified ; during a part of the period T , S is positive and fluctuations tend to grow, but before the Fredericksz transition can occur, S becomes negative and the fluctuations relax [5].

5. **Defects.** — In the above discussions, we have supposed that the distortion was uniform in the whole sample. We know in fact, that, if the magnetic field is exactly normal to \mathbf{n}_0 , there is some degeneracy for the distortion, which is no longer uniform.

In the zero frequency limit, the distortions θ and $-\theta$ are energetically equivalent, and we obtain domains, separated by walls [6-7].

In the infinite frequency limit, there is a complete degeneracy for φ , and we obtain umbilics [8], completely similar to those obtained for the electrical field analogue given in the introduction.

For intermediate frequencies, we obtain some very nice configurations, with spirals, or even some more complicated figures. An example of a spiral is shown on figure 7.

6. **Conclusion.** — We have seen that when a nematic slab is in a magnetic field normal to \mathbf{n}_0 and rotating in the plane of the slab, then its behavior is dominated by two threshold fields :

$H_a(\omega)$ threshold for the synchronous rotation of the director.

$H_c(\omega)$ threshold for the distortion.

For $\omega = \omega_1$, $H_a(\omega) = H_c(\omega)$.

For $\omega < \omega_1$, the distortion appears in the synchronous regime, and looks very similar to the static one, except that the threshold $H_c(\omega)$ is larger than H_{c_0} .

For $\omega > \omega_1$, the distortion appears in the asynchronous regime. The director rotates more slowly than the field. The retardation angle between the field and the director is periodic in time, with a periodicity which diverges as H is increased to $H_a(\omega)$.

Experimental observations are in good agreement with those predictions, and the Fredericksz transition in rotating fields reveals itself a useful tool to investigate not only the static, but also the dynamic behaviour of the nematic mesophases. In particular, the measure of H_a versus ω is an accurate way to determine the twist viscosity γ_1 .

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