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TWO FREEDERICKSZ TRANSITIONS IN CROSSED ELECTRIC AND MAGNETIC FIELDS

H. J. DEULING (*)

Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France

À. BUKA and I. JÀNOSSY

Central Research Institute for Physics, H-1525, Budapest 114, P.O.B 49, Hungary

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Résumé. — Un film nématique planaire présente une transition de Freedericksz dans un champ électrique perpendiculaire aux plaques. Si on applique en plus un champ magnétique parallèlement aux couches (mais perpendiculairement au directeur), deux transitions se produisent à $V = V_s(H)$ et $V = V_t(H)$. Au-dessous de V_s on n'a qu'une torsion du liquide. Au-dessus de V_s, on trouve en plus une distorsion en éventail qui provoque la diminution de la torsion qui disparaît à $V = V_t(H)$. Nous calculons la distorsion entre V_s et V_t et comparons nos résultats à des expériences sur p-p-dibutylazoxybenzène.

Abstract. — A planar nematic slab shows a Freedericksz transition in a perpendicular electric field. Applying in addition a magnetic field parallel to the slab but perpendicular to the direction of alignment one finds two transitions at $V = V_s$ and $V = V_t$. Below V_s one has only a twist deformation. Above V_s splay and bend also appear causing the twist to diminish. At $V = V_t$ the twist disappears. We calculate the distortion between the thresholds and compare our results with data on p-p-dibutylazoxybenzene.

A nematic liquid enclosed between two glass plates can be oriented by proper treatment of the glass surfaces such as to form a liquid monocrystal with axis parallel to the plates. Such a nematic slab can be deformed by external electric or magnetic fields. The resulting distortion is governed by a balance of stabilizing elastic torques and destabilizing torques due to external fields. If a magnetic field is applied perpendicular to the slab a splay and bend distortion is induced when the field exceeds a threshold [1-3] $H_{c_1} = (\pi/L)(K_1/\chi_a)^{1/2}$. In this expression L denotes the thickness of the slab, K_1 is the splay elastic constant and $\chi_a = \chi_\parallel - \chi_\perp$ denotes the anisotropy of the susceptibility. If the magnetic field is applied parallel to the slab but still perpendicular to the direction of alignment, a twist distortion is induced as soon as H exceeds a threshold $H_{c_2} = (\pi/L)(K_2/\chi_a)^{1/2}$ where K_2 is the twist elastic constant. Recently, these two cases have been shown to be only the limiting forms of the more general case [4], when the field forms an oblique angle ψ with the slab but is still perpendicular to the direction of alignment. The threshold is then given by the expression

$$H_c^{-2}(\psi) = H_{c_1}^{-2} \sin^2 \psi + H_{c_2}^{-2} \cos^2 \psi \,. \quad (1)$$

When H exceeds this threshold splay, twist and bend are induced simultaneously.

One obtains two different thresholds for twist and splay, however, if one employs a parallel magnetic field to induce twist and uses a vertical electric field to induce splay and bend. If we turn on the magnetic field first we start with a pure twist distortion. Turning on the voltage, we will induce in addition a splay and bend distortion at some threshold value $V_s(H)$ [5]. If on the other hand, we first apply the voltage we start with a splay and bend distortion. Now we turn on the magnetic field H beyond H_{c_2} but keep H still low enough to induce no twist distortion. If we now decrease the voltage, a twist distortion will appear at some threshold $V_t(H)$.

This conspicuous difference between the Freedericksz transitions in an oblique magnetic field and in crossed electric and magnetic field, respectively, can be understood as follows : the torques per unit volume acting on the liquid due to a magnetic field

(*) On leave from : Freie Universität Berlin, FB 20-WE 05, 1 Berlin-33, Arnimallee 3.

are proportional to $(\mathbf{H}.\mathbf{n})^2$. Decomposing $\mathbf{H}$ into parallel and perpendicular components, we get terms proportional to

$$(\mathbf{H}_\perp.\mathbf{n})^2 + (\mathbf{H}_\parallel.\mathbf{n})^2 + 2(\mathbf{H}_\perp.\mathbf{n})(\mathbf{H}_\parallel.\mathbf{n}).$$

In crossed electric and magnetic fields, however, we get torque densities proportional to $(\mathbf{E}.\mathbf{n})^2$ and $(\mathbf{H}.\mathbf{n})^2$ but there is no coupling term of the kind $2(\mathbf{E}.\mathbf{n})(\mathbf{H}.\mathbf{n})$ as we found for the oblique magnetic field, replacing $\mathbf{E}$ by $\mathbf{H}_\perp$ and $\mathbf{H}$ by $\mathbf{H}_\parallel$, respectively.

For a rigorous solution, we start from the Gibbs free energy G per unit area of the sample. We take the sample to be in the x-y plane of a coordinate system with H along the x-axis and the direction of alignment along the y-axis. We introduce polar angles φ and ω writing the director as

$$\mathbf{n} = (\cos\varphi \sin\omega, \cos\varphi\cos\omega, \sin\varphi). \quad (2)$$

The free energy is then of the form [6, 7]

$$G = (1/2)\int \{ (K_1\cos^2\varphi + K_3\sin^2\varphi)(\mathrm{d}\varphi/\mathrm{d}z)^2 + (K_2\cos^2\varphi + K_3\sin^2\varphi)\cos^2\varphi(\mathrm{d}\omega/\mathrm{d}z)^2 - \chi_a H^2\cos^2\varphi\sin^2\omega\}\,\mathrm{d}z - (8\pi)^{-1} D.V \quad (3)$$

where V is the applied voltage and D the z-component of the dielectric displacement which is a functional of the angle of tilt $\varphi(z)$.

$$D = V\Big/\int_0^L (\varepsilon_\parallel\sin^2\varphi + \varepsilon_\perp\cos^2\varphi)^{-1}\,\mathrm{d}z. \quad (4)$$

Minimizing the free energy G, we get two differential equations of second order for the functions $\varphi(z)$ and $\omega(z)$. We measure the magnetic field in units of H_{c_2}, the voltage in units of $V_{c_1} = 2\pi^{3/2}(K_1/\varepsilon_a)^{1/2}$ and D in units of $D_{c_1} = \varepsilon_\perp V_{c_1}/L$. We also introduce parameters

$$\kappa = (K_3 - K_1)/K_1$$
$$\alpha = (K_3 - K_2)/K_2 \quad (5)$$
$$\gamma = (\varepsilon_\parallel - \varepsilon_\perp)/\varepsilon_\perp.$$

The equations for $\varphi(z)$ and $\omega(z)$ are then

$$\frac{\mathrm{d}}{\mathrm{d}z}\left\{(1+\alpha\sin^2\varphi)\cos^2\varphi\frac{\mathrm{d}\omega}{\mathrm{d}z}\right\} = -(\pi/L)^2(H/H_{c_2})^2\cos^2\varphi\sin\omega\cos\omega$$

$$\frac{\mathrm{d}}{\mathrm{d}z}\left\{(1+\kappa\sin^2\varphi)\frac{\mathrm{d}\varphi}{\mathrm{d}z}\right\} - \kappa\sin\varphi\cos\varphi(\mathrm{d}\varphi/\mathrm{d}z)^2 - [(1+\kappa)/(1+\alpha)]\{\alpha(\cos^2\varphi - \sin^2\varphi) - 1\}\sin\varphi\cos\varphi(\mathrm{d}\omega/\mathrm{d}z)^2 = (\pi/L)^2[(1+\kappa)/(1+\alpha)](H/H_{c_2})^2\sin^2\omega\cos\varphi\sin\varphi - (\pi/L)^2(D/D_{c_1})^2\sin\varphi\cos\varphi(1+\gamma\sin^2\varphi)^{-2}. \quad (6)$$

In the vicinity of $V_s(H)$, the angle $\varphi(z)$ is very small. Expanding the equations to lowest order in φ, we get :

$$\mathrm{d}\omega/\mathrm{d}z = \pm(\pi/L)(H/H_{c_2})\{\sin^2\omega_m - \sin^2\omega\}^{1/2}$$

$$\mathrm{d}^2\varphi/\mathrm{d}z^2 = -(\pi/L)^2\{(D/D_{c_1})^2 - (H/H_{c_2})^2[(1+\kappa)/(1+\alpha)][(\alpha-1)\sin^2\omega_m - (\alpha-2)\sin^2\omega]\}\varphi \quad (7)$$

$$\omega_m = \omega(z = L/2).$$

The equation for φ is an eigen-value equation for the eigen-value $(D/D_{c_1})^2$. The threshold-voltage $V_s(H)$ is obtained from expression (4)

$$V_s(H)/V_{c_1} = (D/D_{c_1})\,L\Big/\int_0^L (1+\gamma\sin^2\varphi)^{-1}\,\mathrm{d}z.$$

In the vicinity of $V_t(H)$, the twist angle $\omega(z)$ is small. Expanding to lowest order in ω, we get

$$\frac{\mathrm{d}}{\mathrm{d}z}\left\{(1+\kappa\sin^2\varphi)\frac{\mathrm{d}\varphi}{\mathrm{d}z}\right\} - \kappa\sin\varphi\cos\varphi(\mathrm{d}\varphi/\mathrm{d}z)^2 = -(\pi/L)^2(D/D_{c_1})^2\sin\varphi\cos\varphi(1+\gamma\sin^2\varphi)^{-2}$$

$$\frac{\mathrm{d}}{\mathrm{d}z}\left\{(1+\alpha\sin^2\varphi)\cos^2\varphi\frac{\mathrm{d}\omega}{\mathrm{d}z}\right\} = -(\pi/L)^2(H/H_{c_2})^2\cos^2\varphi\,\omega. \quad (8)$$

For any given value of D, we solve the first equation and calculate the corresponding voltage from expression (4). Then, we insert the solution $\varphi(z)$ into the equation for ω and determine the eigen-value $(H/H_{c_2})^2$. In this way, we obtain that value of H which is a root of $V = V_t(H)$. In figure 1, we show the two thresholds

$V_s(H)$ and $V_t(H)$ for $\kappa = 0.2$, $\alpha = 1.4$ and $\gamma = 0.24$. The parameters were chosen so as to correspond to p-p-dibutylazoxybenzene for which experimental data are presented below.

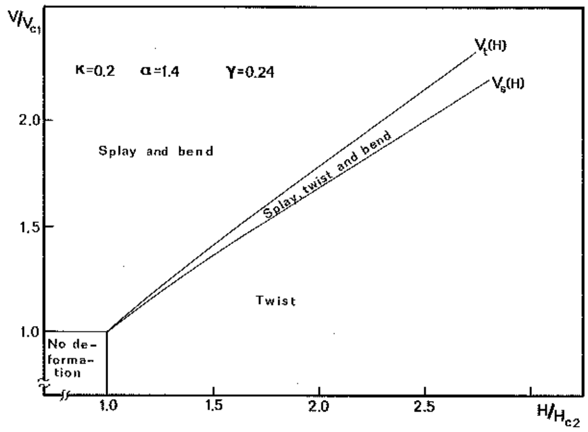

FIG. 1. — Thresholds V_s and V_t as functions of H. The diagram indicates the types of deformation displayed by the sample.

It is of some interest to look at eqs. (7) and (8) in the limit of $H \gg H_{c_2}$. We rewrite eq. (7) in terms of $n_z = \sin \varphi \approx \varphi$. For eq. (8) we make the approximation $n_x = \sin \omega \cos \varphi \approx \omega \cos \varphi$. For strong magnetic fields, we find in the vicinity of $V_s(H)$

$$\omega(z) \approx \omega_m \approx \pi/2$$

nearly everywhere in the sample except in a thin layer near the boundaries where $\omega(z)$ rapidly goes to zero. In this approximation, outside the boundary layer, eq. (7) reads

$$n_z'' = -(\pi/L)^2 \{ (V/V_{c_1})^2 - [(1+\kappa)/(1+\alpha)] (H/H_{c_2})^2 \} n_z \quad (9)$$

n_z is proportional to $\sin(\pi z/L)$ and the threshold $V_s(H)$ is [5]

$$V_s(H) = V_{c_1} \{ 1 + [(1+\kappa)/(1+\alpha)] (H/H_{c_2})^2 \}^{1/2} . \quad (10)$$

This derivation shows that the threshold $V_t(H)$ is influenced mainly by the twist distortion pattern in the middle of the layer.

In the vicinity of the second threshold $V_t(H)$ we have over a wide range in the middle of the sample $\varphi(z) \approx \varphi_m \approx \pi/2$ and $\varphi' \approx 0$. From expression (4), we find $V/V_{c_1} \approx (1+\gamma)^{-1} D/D_{c_1}$. With these approximations, we may rewrite eq. (8) in the form

$$n_x'' = (\pi/L)^2 (1+\kappa)^{-1} \{ (V/V_{c_1})^2 - [(1+\kappa)/(1+\alpha)] (H/H_{c_2})^2 \} n_x . \quad (11)$$

Comparing this equation for n_x with that for n_z (9), we see that the expression in curly brackets in (11) is positive, which is to say that in the middle of the layer $n_x(z)$ is proportional to $\cosh(c.z)$, i.e. $n_x(z)$ has a minimum at $z = L/2$. This shows that the threshold $V_t(H)$ is controlled by the splay and bend distortion in the vicinity of the boundaries.

Having determined the critical voltages we solve the eq. (6) for $\varphi(z)$ and $\omega(z)$ for voltages in the interval $V_s(H) \leqslant V \leqslant V_t(H)$. The distortion is measured by monitoring in the usual way [7, 8] the shift of birefringence δ.

$$\delta . \lambda/(n_e . L) = 1 - (1/L) \int_0^L (1 + \nu \sin^2 \varphi)^{-1/2} \, dz \quad (12)$$

λ is the wavelength of the light, n_e the extraordinary index of refraction and $\nu = (n_e/n_0)^2 - 1$.

On figure 2, we give experimental data obtained on p-p-dibutylazoxybenzene at 25 °C. The data taken in a magnetic field clearly display two thresholds. In a magnetic field of sufficient strength, there is no shift of birefringence below a certain voltage (V_s) while above another voltage (V_t) the shift of birefringence practically equals that obtained for $H = 0$.

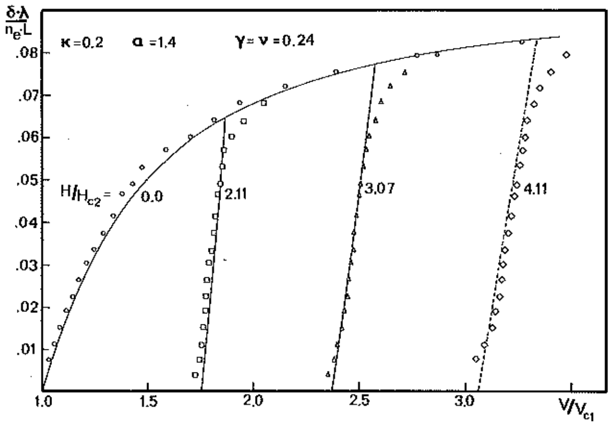

FIG. 2. — Comparison of theoretical calculations of the shift of birefringence δ versus voltage with data on p-p-dibutylazoxybenzene. For high magnetic fields only the thresholds V_s and V_t could be calculated but not the curve in between. For $H = 4.11 \times H_{c_2}$ we have therefore connected the thresholds by a dotted line.

To compare the data with the results of numerical calculations based on eq. (6) we first consider the data obtained in zero magnetic field, which depend only on K_1 and K_3. From data for the dielectric and optical anisotropy, we find $\gamma = \nu = 0.24$. Using this number, the best fit to the data for $H = 0$ is obtained with $\kappa = (K_3 - K_1)/K_1 = 0.2$, that is $K_3 = 1.2\ K_1$.

The data taken in a magnetic field exceeding H_{c_2} depend not only on K_3 and K_1, but on K_2 as well. We found good agreement between the theoretical curves and the data taking $\alpha = (K_3 - K_2)/K_2 = 1.4$, or $K_3 = 2.4\ K_2$, $K_1 = 2\ K_2$. The theoretical curves for $H/H_{c_2} = 2.11$ and 3.07 shown in figure 1 are nearly straight lines connecting the two points corresponding to the thresholds V_s and V_t, respectively. The small round-off in the experimental data near the thresholds is likely to be due to either small misalignments of the director at the boundaries or a small misalignment of the magnetic field relative to the sample. (For $H/H_{c_2} = 4.11$, we could only calculate the thresholds but not the curve in between. We have connected by a dotted line the two points corresponding to V_s and V_t.)

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References

[1] FREEDERICKSZ, V., ZOLINA, V., *Z. Kristallogr.* **79** (1931) 225.
[2] ZOCHER, H., *Z. Physik* **28** (1927) 790.
[3] PIERANSKI, P., BROCHARD, F. and GUYON, E., *J. Physique* **33** (1972) 681.
[4] DEULING, H. J., GABAY, M., GUYON, E. and PIERANSKI, P., *J. Physique* **36** (1975) 689.
[5] DEULING, H. J., GUYON, E. and PIERANSKI, P., *Solid. State Commun.* **15** (1974) 277.
[6] DEULING, H., *Mol. Cryst. and Liq. Cryst.* **19** (1972) 123.
[7] GRULER, H., SCHEFFER, T. and MEIER, G., *Z. Naturforsch.* **27A** (1972) 966.
[8] SAUPE, A. Z., *Z. Naturforsch.* **15a** (1960) 815.