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Dynamics of the Orientation of a Nematic-Liquid-Crystal Film in a Variable Magnetic Field

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The transient distortions, induced in a nematic film by a stepwise increase (or decrease) of the magnetic field around the threshold value H_c, are accurately measured by conoscopic techniques. The results can be interpreted by the Leslie equations and provide a measurement of a viscosity coefficient.

Suppose a single-crystal nematic film anchored on glass boundary surfaces is subjected to a magnetic field H perpendicular to its optical axis: The configuration gets distorted when H is larger than the Freedericksz critical field H_c. The static distortions induced by $H > H_c$ have been studied in detail.[1,2] We present here an experimental study of the dynamic behavior under variable H and what we believe to be the first discussion of it, based on the Leslie theory.[3] Let us first recall the static theory.

If θ is the angle between the unperturbed optic axis $\vec{n}_0$ and $\vec{n}$, the Frank[4] free energy per cm^2 is

$$F = \tfrac{1}{2}\chi_a H^2 \int_{-d/2}^{d/2} dz \times [\xi_i^2(1+\eta_i \sin^2\theta)(d\theta/dz)^2 - \sin^2\theta]. \quad (1)$$

The three configurations studied here ($i = 1, 2, 3$) and the geometrical factors are shown in the inset of Fig. 1; $\xi_i = H^{-1}(K_{ii}/\chi_a)^{1/2}$ is a coherence length; χ_a the anisotropic part of the diamagnetic susceptibility; η_i measures the anisotropy of the elastic coefficients K_{ii} [$\eta_1 = (K_{33}-K_{11})/K_{11}$, $\eta_2 = 0$, $\eta_3 = (K_{11}-K_{33})/K_{33}$]. Assuming strong anchoring [$\theta(\pm d/2) = 0$], the optimum configuration near H_c corresponds to $\theta = \theta_M \cos(\pi z/d)$ and (1) reduces to

$$F = \tfrac{1}{2}\chi_a H^2 \{[(\xi_i \pi/d)^2 - 1]\tfrac{1}{2}\theta_M^2 + (1+\eta_i)\tfrac{1}{8}\theta_M^4\}. \quad (2)$$

We recognize a Landau expansion in the neighborhood of a second-order transition.[5] We set $d/\pi\xi_i = H/H_c = h$. For $h < 1$, the stable state is $\theta_M = 0$; for $h > 1$, it corresponds to

$$\theta_M^2 = 2(1-h^{-2})/(1+\eta_i). \quad (3)$$

We studied homeotropically aligned (geometry 3) methoxybenzilidene butylanilin (MBBA). The dynamic behavior was followed by measuring, between crossed polaroids in a monochromatic light, the motion of fringes which are due to the birefringence when $\theta_M \neq 0$. A typical recording,

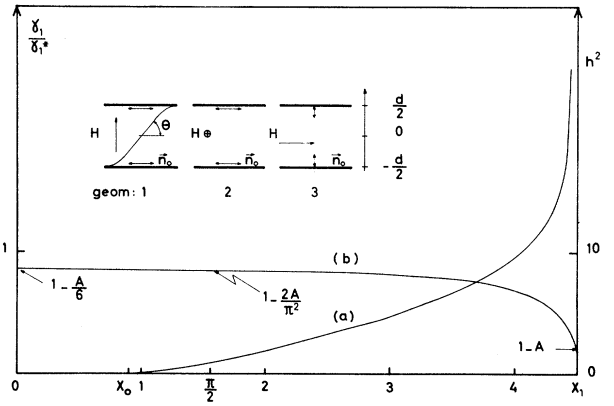

FIG. 1. MBBA in geometry 3: curve a, the wave vector of the distortion $k = 2d^{-1}X$ as a function of the reduced field $h = H/H_c$; curve b, the normalized effective viscosity $\gamma_1^*(X)$. It is practically a constant for the fields values used in this experiment. The inset refers to the three geometries discussed.

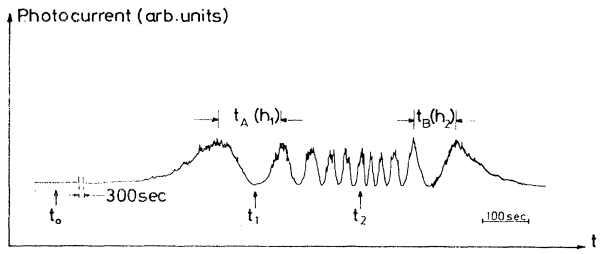

FIG. 2. Dynamical behavior when a field h_1 is applied at time t_0 (or is suddenly decreased to h_2 at time t_2), characterized by the time between the first two successive conoscopic fringes $t_A(h_1)$ [$t_B(h_2)$].

using a small photocell detector, is shown in Fig. 2. A stepwise field $h_1 > 1$ is applied at time t_0 (case A). The distortion, characterized by the number of the fringe N (counted from the undistorted state), increases exponentially. The time constant $s^{-1}(h_1)$ is obtained from the time difference $t_A(h_1) = t_A'' - t_A'$ between two fringes:

$$N(t_A'')/N(t_A') = \exp[s(h_1)t_A(h_1)]. \tag{4}$$

At a further time t_2, the field is decreased to a value $h_2 < 1$ (case B), we consider again the time difference $t_B(h_2) = t_B' - t_B''$ corresponding to fringes such that $N(t_A'')/N(t_A') = N(t_B'')/N(t_B')$. We obtain the relations

$$\frac{s(h_1)}{|s(0)|} = \frac{t_B(0)}{t_A(h_1)}, \quad h_1 > 1;$$
$$\frac{s(h_2)}{|s(0)|} = -\frac{t_B(0)}{t_B(h_2)}, \quad h_2 < 1. \tag{5}$$

The times are normalized to the value for $h_2 = 0$, $t_B(0)$, associated with a decay time $s^{-1}(0) < 0$. In this limit of small distortions, $\theta_M^2(t) = g(h)N(t)$. The constant s^{-1} also describes the exponential variation of $\theta_M^2(t)$.[6] The experimental ratios are given in Fig. 3 for two samples ($d = 180$ and 330 μm). They follow a $h^2 - 1$ law from $h = 0$ to $h = 2$ with a better than 10% accuracy. At $h = 1$, the relaxation times diverge like $|1 - h|^{-1}$. This divergence is a common feature of second-order transitions. The "mean-field" exponent -1 applies here because fluctuations are weak on a macroscopic dimension (ξ_i is large, as in the superconducting problem).

Let us analyze the two behaviors A and B in the three geometries. In cases 1 and 3, the gradient of the angular velocity of $\vec{n}$ produces a backflow motion of the fluid, causing a frictional torque[3] on $\vec{n}$: $\vec{\Gamma}' = \vec{n} \times \gamma_1(-\vec{\omega} \times \vec{n}) + \gamma_2 \vec{A} \cdot \vec{n}$, where $2\vec{\omega} = \text{curl}\vec{v}$ and $2A_{i,j} = (v_{i,j} + v_{j,i})$. For an incompressible

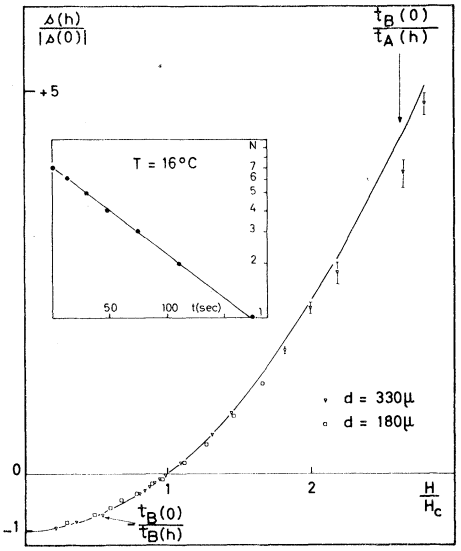

FIG. 3. Time constant of the distortion, as measured by $s^{-1}(h)$, normalized to the value for $h = 0$, which follows the $h^2 - 1$ law predicted accurately by the theory. Note in particular the divergence of the time constant as $|1 - h|^{-1}$ at H_c. The exponential relaxation of the distortion for $h_2 = 0$, close to the equilibrium, is followed by the indexed number of fringes $N(t)$ (inset) and is used to measure γ_1*.

fluid, the only nonzero component of the velocity is $v_x(z, t)$. The equilibrium between elastic, magnetic, and viscous torques is given by

$$\xi_i^2 \frac{\partial^2 \theta}{\partial z^2} + \theta = \tau_0 \frac{\partial \theta}{\partial t} + \lambda_i \tau_0 \frac{\partial v}{\partial z}, \tag{6}$$

where $\tau_0 = \gamma_1/\chi_a H^2$, $\lambda_1 = (\gamma_1 + \gamma_2)/2\gamma_1$, $\lambda_3 = (\gamma_2 - \gamma_1)/2\gamma_1$. Neglecting inertial effects (correct here), $\vec{v}$ is given by the Leslie equation,

$$\frac{\partial}{\partial z}\left(a_i \frac{\partial v_x}{\partial z} + b_i \frac{\partial \theta}{\partial t}\right) = 0, \tag{7}$$

$a_1 = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6)$; $b_1 = \alpha_3$; $a_3 = \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2)$; $b_3 = \alpha_2$. The θ and v_x solutions, satisfying the boundary conditions $\theta(\pm d/2) = v_x(\pm d/2) = 0$, are

$$\theta = \theta_0(\cos kz - \cos \tfrac{1}{2}kd)e^{st}, \tag{8}$$

$$v_x = v_0[\sin kz - (2z/d)\sin \tfrac{1}{2}kd]e^{st}. \tag{9}$$

Equations (6) and (7) lead to the following conditions:

$$(1 - A_i)s\tau_0 = 1 - 4X^2/\pi^2 h^2, \tag{10}$$

$$h^2 = \frac{4X^2}{\pi^2} \frac{\tan X - X/A_i}{\tan X - X}. \tag{11}$$

The constant $A_i = \lambda_i b_i / a_i$ ($0 < A_i < 1$) gives the reduction of the viscosity in an infinite medium due

to the backflow.[7] (Using the viscosity coefficients of MBBA,[8] one gets $A_1 = 10^{-3}$, $A_3 \sim 0.75$.) The results of the coupled equations (10) and (11) are given in Fig. 1 for geometry 3. The factor $X = kd/2$ (curve a) gives the wave vector of the distortion compared with the static value $k = \pi/d$. The fastest increase (case A) and the slowest decay (case B) of θ_M, which are seen in the experiments, are both given by the smallest X solution [see Eq. (10)] which corresponds to the weakest distortion. In geometry 2, where there is no backflow ($A_2 = \lambda_2 = 0$), the time constant $s_0^{-1}(h)$ can be calculated directly from (6);

$$s_0(h) = \chi_a H_c^2 (h^2 - 1)/\gamma_1. \quad (12)$$

The variation of $s(h)$ is plotted in terms of the ratio (curve b)

$$s_0(h)/s(h) = \gamma_1^*(h)/\gamma_1. \quad (13)$$

The variation of the effective viscosity $\gamma_1^*(h)$ should not be observable in our experiments where $0 < h < 2$. We thus expect, using (12) and (13),

$$\frac{s(h)}{|s(h=0)|} = \frac{\chi_a (h^2 - 1) H_c^2}{\gamma_1^*} \frac{\gamma_1^*}{\chi_a H_c^2} = h^2 - 1,$$

as was accurately obtained in the experiments, shown in Fig. 3.

Finally, we determine an absolute value of γ_1^* from the exponential variation of N versus t when a field is suppressed (see inset of Fig. 3). Using $\chi_a \simeq 1.2 \times 10^{-7}$ and the correction $\gamma_1 = 1.16\gamma_1^*$ (see Fig. 1, curve b), we get $\gamma_1 = 1.95$ at 16°C and $\gamma_1 = 1.25$ at 22°C, in agreement with other temperature-dependent results.[9] More detailed results and data for geometry 1 will be given elsewhere.[2]

It is a pleasure to acknowledge critical discussions with P. G. de Gennes and L. Léger.

[1]See, for example, H. Zocher, Trans. Faraday Soc. 29, 945 (1933).

[2]P. Pieranski, F. Brochard, and E. Guyon, to be published.

[3]We use the notations of Groupe d'Etudes des Cristaux Liquides, J. Chem. Phys. 51, 816 (1969).

[4]F. C. Frank, Discuss. Faraday Soc. 25, 1 (1958).

[5]L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, London, 1959), p. 430.

[6]In case A, $\theta_M^2(0)$ is the mean square value of the thermal fluctuations which induces a transition from a maximum of F for $\theta = 0$ to the equilibrium state. The equipartition theorem leads to $\theta_M^2(0) \sim a/d$, where a is a molecular dimension. Other contributions come from residual misalignment of H or in the film (Ref. 2).

[7]P. Martinoty and S. Candau, Mol. Cryst. Liquid Cryst. 14, 243 (1971).

[8]C. Gähwiller, Phys. Lett. 36A, 311 (1971).

[9]J. Prost and H. Gasparoux, Phys. Lett. 36A, 255 (1971) (cgs units are used).

Solid Surface Shape and the Alignment of an Adjacent Nematic Liquid Crystal

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We show that elastic strain energy may account for the tendency of some nematic liquid crystals to lie parallel to the direction of rubbing on a solid surface that has been slightly deformed by rubbing, or perpendicular to a surface that is slightly rough in two dimensions.

It is well known that the directors of many nematic liquid crystals tend to assume an orientation parallel to the direction in which an adjacent solid surface has previously been rubbed. Such rubbing may be done on glass with fresh dry lens tissue or cloth, or with a polishing lap charged with jeweler's rouge or other polishing material. Dreyer[1] reported that parallel alignment sometimes occurs even in replicas of rubbed surfaces. This observation suggests that one mechanism for such alignment is based primarily on geometrical factors rather than detailed molecular forces. A simple explanation of orientation parallel to the rubbing direction can be made on the basis of the additional elastic energy that would occur in a nematic liquid crystal due to distortion near a gratinglike wavy surface if the liquid crystal molecules were forced to lie against the surface with directors lying across, rather than parallel to, the grooves and ridges. If the surface is rough in both dimensions, such energy considerations would explain a tendency for the molecules