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AZIMUTHAL REORIENTATION OF HOMEOTROPIC NEMATIC FILMS

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Abstract Homeotropic films of a nematic liquid crystal with negative dielectric anisotropy were deformed by an electric field, in the presence of a transverse magnetic field. After removing the magnetic field, azimuthal reorientation of the director was observed. The kinetics of the reorientation process was investigated under various circumstances. The observations can be interpreted assuming a small pretilt of the director at the boundaries.

INTRODUCTION

In this paper, homeotropically aligned films of a nematic liquid crystal with negative dielectric anisotropy are considered. As it is well known, when electric field is applied to such layers, the director configuration becomes deformed above a threshold voltage (electric Freedericksz transition). In the distorted state, the variation of the polar angle of the director along the cell normal can be readily determined by considering the minimum of the total (dielectric + elastic) free-energy or, alternatively, the balance between the dielectric and elastic torques[1]. On the other hand, it is not clear what factors determine the *azimuthal* angle of the director above the Freedericksz threshold. In an ideal case, the free-energy would be invariant with respect to rotations around the surface normal, therefore the azimuthal angle could be arbitrary. In real samples, however, one finds systematically that there exists an "easy axis" for the **c** director (i.e. the unit vector, parallel to the projection of the director onto the substrates).

Consider a sample, on which a voltage, exceeding the threshold, is applied in a stepwise manner. At the beginning, a network of defects (umbilics[2]) is formed. The umbilics represent singularities of strength of ± 1 in the distribution of the **c** director.

This texture, however, is unstable; neigbouring umbilics with opposite signs attract and evantually annihilate each other. In the stabilized structure, defects occur only at the edges of the cell, otherwise the **c** director varies smoothly within the substrate plane.

Experiments show that while umbilics form in a random way (i.e. each time the voltage is switched on, a different network is obtained), the relaxed structure is always the same. The equilibrium distribution of the **c** director is therefore a characteristic of the sample.

The aim of the present work is to investigate quantitatively the relaxation process of the **c** director towards its equilibrium state. In our experiments, the electric field was switched on in the presence of a magnetic field, aligned parallel to the substrates. Using this method, uniform initial conditions of the **c** director were produced. At a certain moment, the magnetic field was removed and the subsequent relaxation was followed by optical detection. Measurements were carried out as a function of the applied voltage. We also compared samples with different geometrical parameters and investigated the effect of prolonged application of the electric field.

In order to interpret the observations, we suggest that there are small deviations from the ideal homeotropic surface alignment in the samples. We calculated theoretically the relaxation process, assuming different anchoring conditions. Satisfactory agreement with the experimental data was obtained assuming strong anchoring and a typical permanent pretilt of a tenth of a degree.

EXPERIMENTAL

In the experiments conventional sandwich-type cells were used. The substrates were commercial indium-tin-oxide coated glasses, with a surface resistivity of $\approx 50\Omega$. In some cells, the conductive layers had been partially removed in such a way that electric field could be applied only on three, well-separated circular regions with a diameter of 2 mm.

We studied the mixture *ZLI 1623* from MERCK, which has a nematic phase from well below the room temperature to 86^0C . The homeotropic alignment was achieved using the traditional method of treating the substrates with lecithin or

a silane compound (octadecyl-trimethoxy-sylane)[3]. The dielectric anisotropy of the material is $\varepsilon_a = -0.6$. The Freedericksz transition was induced by an electric field of a few kHz; this frequency was high enough to avoid electrohydrodynamical instabilities. The threshold voltage was found to be $5.7V$. All measurements were carried out at room temperature.

In a typical experiment, the voltage was switched on in the presence of a magnetic field of $\approx 1kG$, that ensured an initially uniform orientation of the **c** director in the whole cell. (In order to prevent formation of domains with +**c** and -**c** directions, the magnetic field was slightly tilted with respect to the substrate planes.) A short time (few minutes) was allowed for the stabilization of the initial configuration, then the magnetic field was switched off. The relaxation of the **c** director was followed in a polarizing microscope, setting the polarizer parallel, the analyzer perpendicular to the initial **c** director. For quantitative measurements, the eyepiece was replaced by a photodetector. Within the monitored area (about 1mm^2) the **c** director was more or less, although not entirely, uniform during the relaxation process.

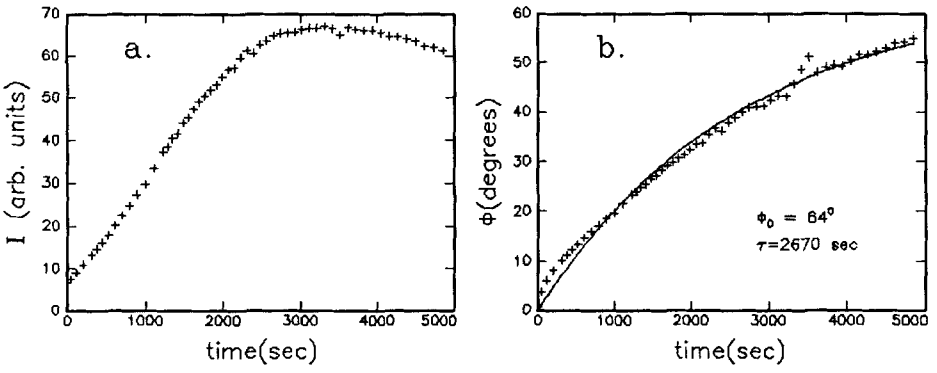

FIGURE 1. *a:* Detected intensity as a function of time. *b:* The reorientation angle, ϕ, as a function of time. The continuous line shows the exponential fit corresponding to Eq. 2.

An example of the recorded curves is shown in Fig. 1a. At $t = 0$, i.e. at the moment when the magnetic field is removed, in principle, no light should pass through the analyzer. The small signal detected in this case (see the figure) is

probably due to the fact that the illuminating light beam was not perfectly parallel. The intensity measured at $t = 0$ was regarded as a background and was subtracted from the further data.

In the general case, as a simple consideration shows, the detected light intensity can be given as

$$I = I_0 \sin^2 2\phi. \tag{1}$$

ϕ is the angle included by the polarizer and the **c** director, I_0 is a constant depending on the sample thickness, the applied electric field and the wavelength distribution of the light-source of the microscope. I_0 can be determined by rotating both the polarizer and the analyzer with 45 degrees and measuring the transmitted intensity in the presence of the magnetic field. Alternatively, one can identify it with the maximum of the detected intensity vs. time curve. The latter method usually yields a slightly smaller value for I_0 than the former one; the difference is caused by the spatial inhomogenity of the **c** director during relaxation. Normally, we applied the second method, because it involves an automatic averaging over the non-uniform distribution of the **c** director within the monitored area. In Fig. 1b. the ϕ angle is shown as a function of time, converted from the data of Fig. 1a, with the help of Eq. 1 and following the procedure outlined above.

RESULTS

In most cases, a reasonable exponential fit of the form

$$\phi = \phi_0(1 - e^{-t/\tau}) \tag{2}$$

could be found for the time dependence of the ϕ angle (Fig. 1b). A deviation of the experimental points from the exponential fit for small t was observed in a number of samples, but not systematically. It should be noted also that, in the case of $\phi_0 > 90^0$, the exponential fit broke down as ϕ approached 90 degrees. According to Eq. 1, $I(t)$ should become zero at $\phi = 90^0$. In reality, due to the spatial inhomogenity of the **c** director, only a minimum was observed. In such cases, the estimation of ϕ_0 was not reliable, therefore we characterized the relaxation time by τ_{max}, i.e. the time belonging to the maximum value of I.

The relaxation time varied significantly from sample to sample, even when the geometrical parameters and the preparation method were virtually identical. The variation was as large as a factor of two. In spite of the large fluctuations in the data, some tendencies could be established unambigously, which are summarized below.

a. Lateral boundary conditions have no significant effect. As mentioned earlier, we prepared cells in which the electrodes on the two plates overlapped only on circular areas with a diameter of 2 mm. The reorientation process in these areas was essentially the same than in samples where the electric field acted in the whole film (typically a rectangular area with a dimension of 10×15 mm).

b. The relaxation time strongly increases with the layer thickness. This effect was studied for example in a wedge-shaped sample (wedge angle 0.25^0) with three circular regions. The layer thicknesses at the centre of the regions were 12, 24 and $36\mu m$; the corresponding relaxation times were 112, 765 and 1208 sec respectively. We note that no systematic differences were found in the relaxation time when a cell with parallel boundaries (wedge angle $< 0.02^0$) and the appropriate part of a wedge-shaped sample, having the same thickness, were compared. The insensitivity to the wedge angle shows that the "geometrical anchoring" effect[4] does not play a role in our case.

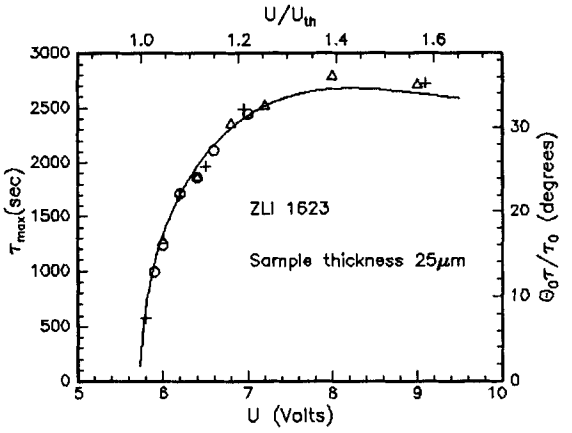

FIGURE 2. τ_{max} as a function of the applied voltage. The scales at the right and the top axes refer to the theoretical curve (solid line). The fit was obtained with $U_{th} = 5.74V$ and $\tau_0/\theta_0 = 77.4$ sec/degrees.

c. The relaxation time as a function of the applied voltage is shown in Fig. 2. As it can be seen from the figure, well above the threshold the relaxation

time depends only weakly on the voltage, but it drops sharply near the threshold. The data were taken on the same sample and reproduced in different runs.

d. During measurements with relatively high voltage ($> 10V$), it was noticed that after long (several hours) application of the electric field, the relaxation time decreased while the maximum of the ϕ vs. time curve somewhat increased. The latter fact indicates a more uniform **c** director distribution. The change in the relaxation process was reversible; after a few hours without applying field, the original relaxation curve recovered (Fig. 3).

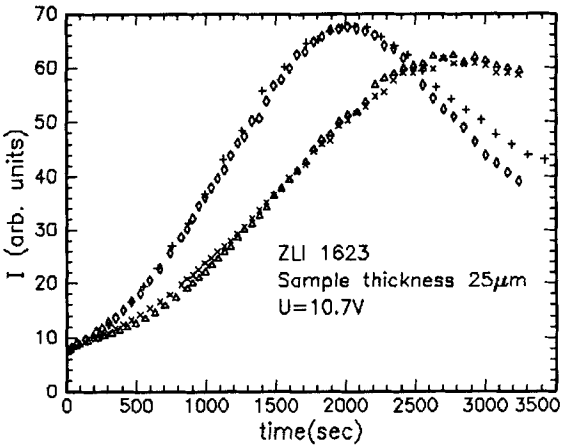

FIGURE 3. Effect of prolonged application of an electric field on the relaxation process. ×: the initial $I(t)$ curve. ◇: the $I(t)$ curve after applying the field for 1 day. +: the $I(t)$ curve after applying the field for 2 days. △: the $I(t)$ curve, 2 days after removing the field.

INTERPRETATION

To account for the observations, we assume that there are small deviations from the perfect homeotropic boundary conditions in the samples. The **n** director at the surfaces has a polar angle of θ_0 and an azimuthal angle of ϕ_0, the latter determing the easy axis of the **c** director.

In an "adiabatic" approximation, the polar angle of the **n** director, θ can be regarded constant during the relaxation process. In the one-constant approximation, the spatial distribution of θ is obtained form the equation[1]

$$K\frac{d^2\theta}{dz^2} - \frac{1}{2}\varepsilon_0\varepsilon_a E^2 \sin 2\theta = 0, \tag{3}$$

subjected to the boundary conditions: $\theta = \theta_0$ at $z = 0$ and $z = L$ (L: sample thickness).

The azimuthal relaxation can be determined from the balance of the elastic and viscous torques:[5]

$$-\gamma_1 \sin^2\theta \frac{d\phi}{dt} + K\frac{d}{dz}\sin^2\theta\frac{d\phi}{dz} = 0 \tag{4}$$

with the boundary conditions $\phi = \phi_0$ at $z = 0$ and $z = L$. Here γ_1 is the rotational viscosity.

The solutions of Eq.(4) are of the form:

$$\phi(z,t) = \Phi(z)e^{-t/\tau} + \phi_0 \tag{5}$$

where Φ obeys the equation

$$\frac{d^2\Phi}{dz^2} + 2\cot\theta\frac{d\theta}{dz}\frac{d\Phi}{dz} + \tau_0/\tau\frac{\Phi}{L^2} = 0 \tag{6}$$

and the boundary conditions:

$$\Phi(0) = \Phi(L) = 0, \tag{7}$$

with $\tau_0 = \gamma_1 L^2/K$.

We determined by numerical calculation the lowest τ_0/τ value at which Eq. 6 has a non-trivial solution compatible with the restrictions given by relation 7. A characteristic feature of this solution is that Φ varies considerably only near the boundaries, where θ is close to zero. It should be noted that there exist further solutions with τ being comparable to τ_0 or smaller; these "fast" modes are, however, important only in the first few seconds of the relaxation process.

It was found from the calculations that τ is a linear function of $1/\theta_0$ for $\theta_0 < 2^0$. The solid curve in Fig. 2 represents the dependence of $\theta_0\tau/\tau_0$ on the applied electric field. An excellent fit with the experimental data is obtained with $\tau_0/\theta_0 =$ 77.4 sec/degrees. Assuming the typical values, $\gamma_1 = 1$ poise and $K = 10^{-11}$ N, one obtains (for $L = 25\mu$m) $\tau_0 \approx 6$ sec. This means a pretilt of

$$\theta_0 \approx 0.08^0.$$

Note that such small pretilts cannot be easily observed using traditional methods (e.g. conoscopic technique) for checking the homeotropic alignment.

According to the theoretical considerations presented here, the relaxation time should be proportional to the square of the thickness. The experimental results, cited earlier are compatible with this prediction.

The model presented here is also compatible with the observation that the rotation of the **c** director can be larger than 90 degrees. As the **c** director is a polar vector, (in contrast with the **n** director, which is an axial vector) the angle included by the initial and final **c** director (i.e. ϕ_0) can be any value between 0 and 180 degrees.

In the above considerations, strong surface anchoring was assumed for both the polar and the azimuthal angle. For the polar angle, the good agreement between the calculated and measured voltage-dependence of the relaxation time justifies this supposition. As a simple analysis shows, in the case of weak polar anchoring, the relaxation time should have a finite value at the threshold and should increase far from the threshold. Such a behaviour is clearly in variance with the experimental results.

The exponential time dependence of ϕ on time indicates that the anchoring is strong for the azimuthal direction too. Deviations from the exponential law at the beginning of the relaxation process may indicate finite azimuthal anchoring energy, but for the time being, our data are not accurate enough to carry out a reliable analysis of this question.

The variation of the relaxation time as the result of prolonged application of an electric field, indicates a plastic behaviour of the surface layer. A similar effect was described recently by Zhuang and Clark[6] for a smectic liquid crystal.

CONCLUSIONS

In this work we demonstrated that in homeotropic layers, prepared by traditional methods, there may be a small pretilt of the order of a tenth of a degree. The magnitude and the azimuthal direction of the pretilt might depend on some fine local characteristics of the substrates, which were not controlled in our sample preparation procedure. Further research is necessary to identify the important factors, determining the pretilt. As a first attempt in this direction, we established that there is no correlation between the direction of the flow during cell-filling and the resulting

azimuthal easy axis.

In most cases, the observed small pretilt can be neglected, but it may play an important role in processes which involve azimuthal reorientation of a distorted director configuration. Examples of such cases are pattern formation in rotating magnetic field[7] and director rotation induced by circularly polarized light[5].

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