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SPATIALLY RESOLVED STUDY OF THE FRÉEDERICKSZ TRANSITION IN THIN NEMATIC CELLS

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Abstract We present a novel method for spatially resolved measurements of the amplitudes of thermal fluctuations and surface pretilt angles in thin cells filled with nematic liquid crystals (LC).

We use the idea that the dynamical response of the director field during a switching process critically depends on the initial conditions of the director field which vary over the cell plane. The applied switching voltage acts as an amplifier which augments initial director deformations by orders of magnitude. By means of a CCD line scan camera combined with a polarizing microscope, we study the time dependence of the optical transmission curves. Their slopes give information on viscoelastic parameters whereas their delay can be related directly to the initial director angle.

INTRODUCTION

The dynamical response of a nematic LC contained in a thin cell to an applied voltage depends not only on the viscoelastic properties but also on the initial orientation of the director field. In planar cells with an electric field normal to the cell planes, the influence of initial deformations on the director field is critical. These initial deviations from the strict planar orientation include pretilt and thermal fluctuations. For the determination of thermal fluctuations, one can use light scattering experiments and NMR methods¹.

For the measurement of pretilt angles in thin cells, the crystal rotation method is accepted in general to yield very precise results ($\pm 0.1^\circ$)^{2,3}. The conoscopic method⁴, the capacitive method and the magnetic null method are further approaches⁵. However, all of these experiments detect an integral value over the cell.

By means of a CCD line scan camera mounted on a polarizing microscope, we have recorded the temporal change of transmitted light after application of an external electric field simultaneously at different local positions along a certain cross section of the cell plane⁶. Corresponding to the local initial conditions of the director field, the individual transmission curves are mutually shifted in time. Theoretical curves are fitted to the experimental variation of transmitted light. From the fit parameters, we determine the initial director deformations in the moment $t = 0$ consisting of pretilt and thermal fluctuations. The switching process acts as an amplifier which augments the initial conditions by orders of magnitude in the direction of the electric field.

THEORY

The optical investigations base on the relations between the director deformation in the cell and the optical phase difference between ordinary and extraordinary waves which are brought to interference at the analyzer. The experiments are performed with crossed polarizers at 45° orientation to the surface director. For small director deformations, geometrical optics can be applied. The transmitted intensity I_A is modulated by

$$I_A(x, y, t) = I_0 \sin^2(\pi N(x, y, t))$$

where the normalized order of interference $\tilde{N}(x, y, t) = (N_{max} - N(x, y, t))/N_{max}$ is related to the director tilt angle amplitude $\theta_m(x, y, t)$ by

$$\tilde{N}(x, y, t) = \sigma \theta_m^2(x, y, t)$$

$$\text{with } N_{max} = \frac{d(n_e - n_o)}{\lambda} \quad \text{and} \quad \sigma = \frac{(n_e + n_o)n_e}{4n_o^2} .$$

θ being the angle formed by the director in the x, z plane with the x axis. The coordinates x, y are in the cell plane while z is normal to the glass plates. This approximation is valid for small tilt amplitudes, and a sine deformation in z direction with maximum tilt θ_m is assumed. n_e, n_o are the principal refractive indices, λ is the light wave length and d is the cell thickness.

If an electric field is switched on to a voltage above the Fréedericksz field, then the tilt angle increases according to⁸

$$\theta_m^2(x, y, t) = \theta_m^2(\infty) \left[1 + \left(\frac{\theta_m^2(\infty)}{\theta_m^2(x, y, 0)} - 1 \right) e^{-\frac{2t}{\tau}} \right]^{-1} \quad \text{with} \quad \tau = \frac{\gamma_1 d^2}{\pi^2 K_1} \frac{U_c^2}{U^2 - U_c^2} . \quad (1)$$

Eq. 1 holds for $\theta(t \rightarrow \infty) \ll 1$, U is the applied voltage and U_c is the Fréedericksz threshold. $\theta_0^2(x, y) = \theta_m^2(x, y, 0)$ includes contributions of the pretilt and thermal fluctuations. Eq. 1 can be written in the form

$$\theta_m^2(x, y, t) = \theta_m^2(\infty) \left[1 + e^{-\frac{2(t-t_0)}{\tau}} \right]^{-1} \quad \text{with} \quad t_0 = \frac{\tau}{2} \ln \left(\frac{\theta_m^2(\infty)}{\theta_0^2(x, y)} - 1 \right) .$$

It is easily seen that differing initial orientations θ_0 lead to a shift t_0 of the reorientation curves in time which is detected optically. We assume that for small tilt deformations, the interactions between adjacent positions in the sample are much smaller than the electric torque and the deformations normal to the cell. The averaged value $\langle \theta_0 \rangle$ is always zero for thermal fluctuations. It gives the mean pretilt angle. Below, we will determine $|\theta_0| = \sqrt{\theta_0^2}$.

The director reorientation and consequently the optical interference during the switching process to a fixed final voltage depend upon the two parameters $\theta_0(x, y)$ and τ . As τ is a function of the LC material parameters and the electric torque, it should be independent of the location in the cell. This is confirmed by the fits in the experiments discussed below. Thus, the spatial inhomogeneities of the switching process detected by the camera can be attributed to varying initial conditions of the director

orientation at the start of the switching process. These initial director deformations can be either thermally activated fluctuations or surface pretilt.

Strictly, for a non-zero pretilted cell no Fréedericksz threshold exists at all, but in practice for small pretilts a 'smeared out' threshold is observed and is determinable⁹. The application of a subcritical destabilizing field prior to switching to a voltage above U_c increases the thermal fluctuation amplitudes as well as the deformation related to pretilt. For the description of this amplification of fluctuation amplitudes in dependence of the subcritical initial voltage U_{pre} , we use a simplified model. Only the ground fluctuation mode along z is considered. Its distortion energy per volume is¹⁰:

$$\mathcal{F} = \frac{1}{2} K_1 \theta_z^2 - \frac{1}{2} \Delta \epsilon \epsilon_0 (U/d)^2 \sin^2 \theta \quad .$$

We equate this energy to $1/2 k_B T$ in the sample volume V and obtain

$$\langle \theta_0^2 \rangle = \frac{2 k_B T d^2}{V \Delta \epsilon \epsilon_0} \frac{1}{U_c^2 - U_{pre}^2} = const \frac{1}{U_c^2 - U_{pre}^2} \quad . \quad (2)$$

EXPERIMENT

The measurements presented in this paper have been obtained with commercially manufactured cells with and without pretilt. Their advantage is the uniform thickness as well as the very exact and uniform surface coating.

In case of the non-pretitled sample, we used a $7 \mu m$ thick cell (Jenoptik) with striped ITO-electrodes (width and gap $100 \mu m$) on the upper glass plate and a plane electrode on the opposite glass plate. We have used these cells because they provided excellent planar alignment. The stripe shape of the electrodes is not relevant for our experiments as we have chosen a cross section far from the electrode edges along the stripes where we can neglect electrode boundary effects. SiO_x ($1 < x < 2$) oblique evaporation provides a planar alignment with practically non-pretilt. The glass plates were mounted together with antiparallel directions. The director orientation of this sample is parallel to the striped electrodes.

A pretilt angle in the second cell has been provided by polyimide coating and rubbing. We have used a cell of $19.6 \mu m$ thickness and non-zero pretilt (LINKAM), antiparallely mounted and with plane electrodes.

Both cells have been filled with PCH5 (Merck), a substance with a positive dielectric anisotropy¹¹ ($\Delta \epsilon = 12.1$) and the elastic constants $K_1 = 8.5 pN$, $K_2 = 5.1 pN$ and $K_3 = 16.2 pN$ at temperature $T = 30.3^\circ C$.

Between crossed polarizers in 45° orientation to the surface alignment, the cells were illuminated with parallel monochromatic light ($\lambda = 578 nm$) normal to the cell plane. The transmission was recorded by a 512 element CCD line scan camera mounted on a polarizing microscope. The maximum spatial and temporal resolutions were $0.7 \mu m$ and $0.5 ms$, resp. The data transfer to the PC is in the μs range, the temporal resolution is restricted by exposure time, the limit of spatial resolution is given by the optics of the microscope. The camera line can be fixed in arbitrary orientations to the surface director alignment. We have recorded the cross sections parallel

and perpendicular to the surface director. The AC (1kHz) voltage at the electrodes could be switched between arbitrary amplitudes synchronized with the frame grabber scanning the camera output (see Fig. 1).

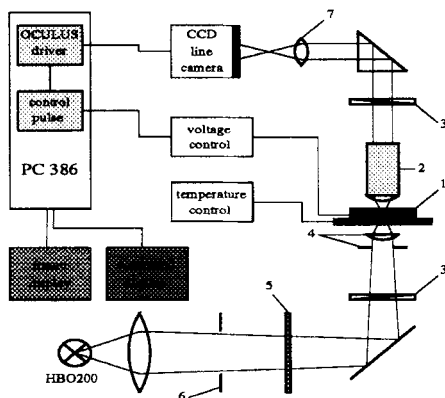


Figure 1: *The experimental setup: 1 sample; 2 objective; 3 polarizers; 4 condensor and aperture; 5 filter; 6 diaphragm; 7 projection lens*

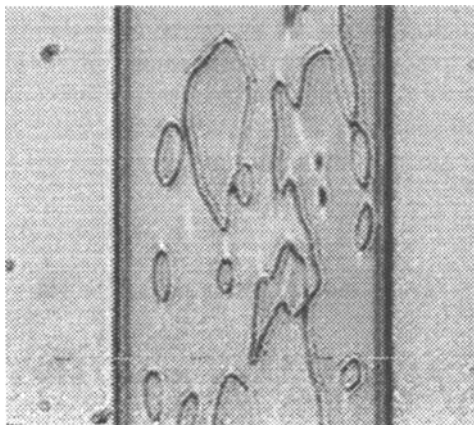


Figure 2: *Domains of opposite tilt in the $7\mu\text{m}$ cell with structured electrode. The image is a top view in the xy plane. The electrode strip is $100\mu\text{m}$ broad and its boundaries appear as vertical dark lines.*

RESULTS

In case of the non-pretilted cell, the response of the nematic director to an applied electric field is retarded because it is in an instable equilibrium at first and the torque of an electric field at 90° to the planar director is zero. Small thermal fluctuations initiate the switching process whereby the director reorientations with $\theta_0 < 0$ and $\theta_0 > 0$ are equivalent. For this reason, domains of opposite tilt may be formed after the switching on process (see Fig. 2). The appearance of these domains indicates that a possible pretilt is much smaller than the thermal fluctuations in the SiO_x cell. In case of the pretilted cell, the nematic switches fast and homogeneously. Pretilt effects can be distinguished from thermal fluctuation effects since a non-zero pretilt usually leads to the preference of one sense of director reorientation, i.e. practically no inversion walls separating domains with opposite tilt are found (see Fig. 8).

Figure 3a shows a transmission pattern obtained from the $7\mu\text{m}$ cell with striped electrodes. The direction of the camera line (space coordinate) was parallel to the director orientation and the direction of electrode stripes, resp. The applied voltage was switched from $U = 0\text{V}$ to $U = 0.9\text{V}$. The critical voltage of Fréedericksz threshold is $U_c = 0.83\text{V}$. In this switching process, the order of interference decreases from $N = 1.3$ (maximum of transmission at $N = 1.5$) to $N = 1.1$ (minimum of transmission at $N = 1$). In dependence on the position along the cross section, the amplitudes of thermal fluctuations vary, leading to a space-dependent U et al. shift of transmission curves and constant switching times. The calculated amplitudes of these fluctuations

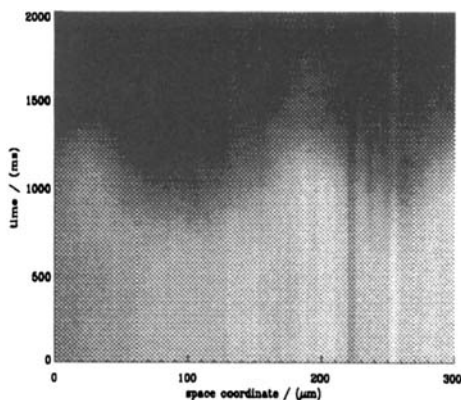


Figure 3a: Transmitted light of the SiO_x $7\mu\text{m}$ cell (no pretilt) after application of a voltage $U = 0.9\text{V}$. The horizontal axis is a cross section along the direction of surface alignment and vertically the time axis is shown.

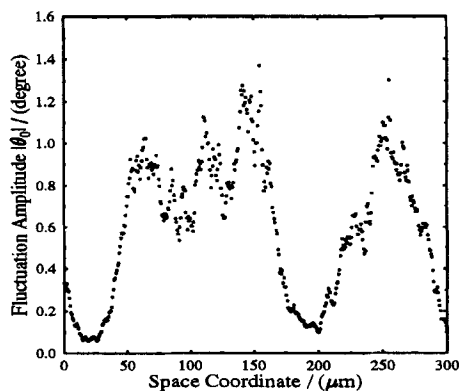


Figure 3b: The calculated values of thermal fluctuations at the moment $t = 0$. The undulations in Fig. 3a are caused by these inhomogeneous initial conditions of tilt angle.

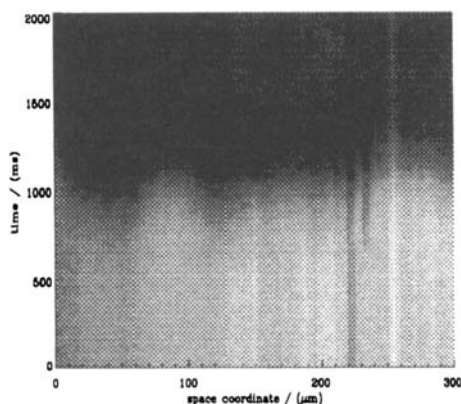


Figure 4a: Same as Fig. 3a, at another switching process. The thermal fluctuations are different but roughly of the same amplitude.

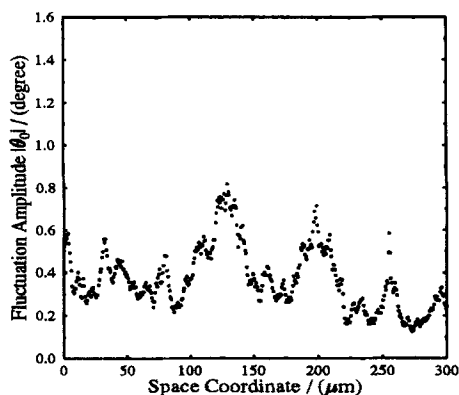


Figure 4b: The calculated values of fluctuations amplitudes from Fig. 4a (cf. Fig. 3b).

are plotted in Figure 3b.

In Figure 4a, another switching process of the same cross section as in Fig. 3 is pictured and in Figure 4b, the calculated amplitudes of fluctuations. In Figs. 3a, 4a, the director field tilts uniformly in one tilt sense, i.e. $\theta < 0$ or $\theta > 0$, which can not be distinguished in our experiments. Very often, we obtained images with inversion walls which separate domains of opposite tilt directions (see Fig. 5). Such images

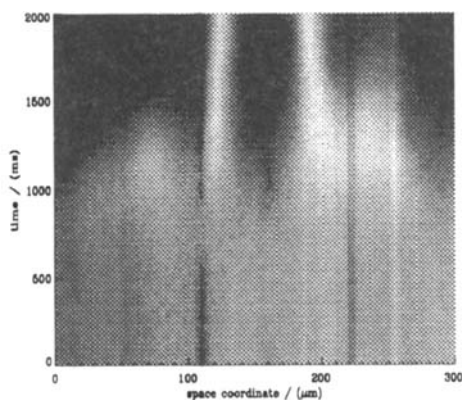


Figure 5: Domains of opposite tilt are separated by inversion walls. These walls appear as vertical bright lines. This image was obtained from the same cross section as Figs. 3 and 4.

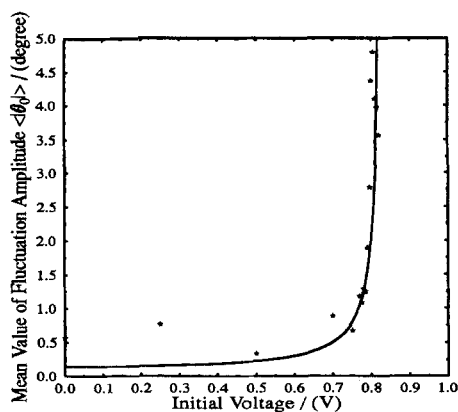


Figure 6: The increase of average $\langle |\theta_0| \rangle$ with increasing subcritical voltage is shown. The solid line is a fit to Eq. 2.

are not suitable for the experimental interpretation. In the vicinity of such walls, the director field is strongly deformed along x and the reorientation of adjacent regions is strongly coupled. Moreover, the appearance of twist near these walls influences the optical transmission¹¹.

Measurements of the averaged thermal fluctuations $\langle |\theta_0| \rangle$ in dependence upon an applied destabilizing voltage U_{pre} ($U_{pre} < U_c$) prior to switching to a voltage $U > U_c$ ($U = 0.9V$, $U_c = 0.83V$) yield the curve depicted in Figure 6. The solid line is a fit to Eq. 2.

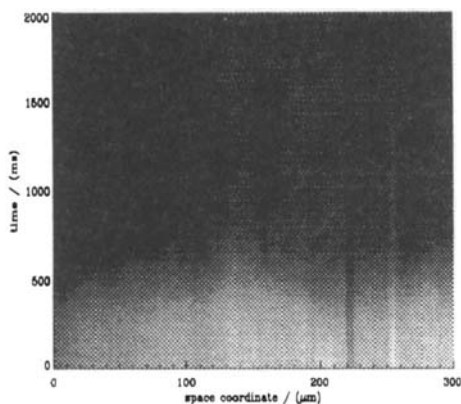


Figure 7a: The interference pattern of a switching process from the voltage $U_{pre} = 0.8V \rightarrow U = 0.9V$ of the same spatial cross section as in Figs. 3 and 4. Note the decreased delay.

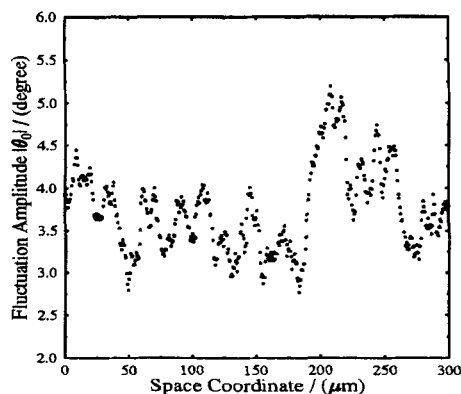


Figure 7b: The calculated fluctuation amplitudes from Fig. 7a. Corresponding to the higher average $|\theta_0|$, the director response to the applied field is faster.

In connection with Eq. 2, Fig. 6 proves the reliability of our method of determination of spatially resolved thermal fluctuation amplitudes.

Figure 7a shows a transmission pattern in the switching process from $U_{pre}=0.8V$ to $U=0.9V$ and Figure 7b the calculated values of fluctuation amplitudes.

For the determination of the pretilt angle in thin cells, we have used the same approach as above. Figure 8a shows the transmission pattern and Figure 8b the calculated $\langle |\theta_0| \rangle$ of the $19.6\mu m$ pretilted cell. The voltage was switched from $U=0V$ to $U=0.9V$.

The spatially resolved values of the pretilt are obtained by averaging the results of

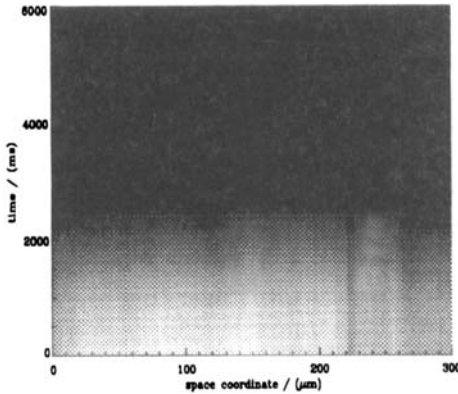


Figure 8a: The interference pattern of a switching process of the $19.6\mu m$ polyimide (pretilted) cell. This cell switches homogeneously and without delay. (Note in Eq. 1, $\tau \propto d^2$.)

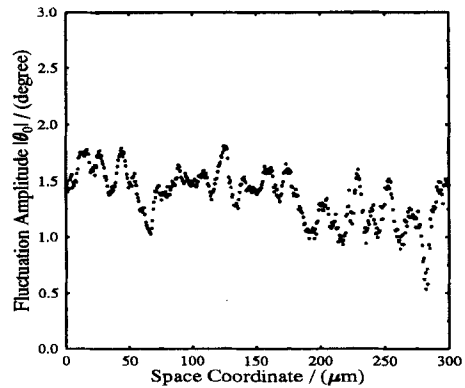


Figure 8b: The calculated fluctuation amplitudes from Fig. 8a.

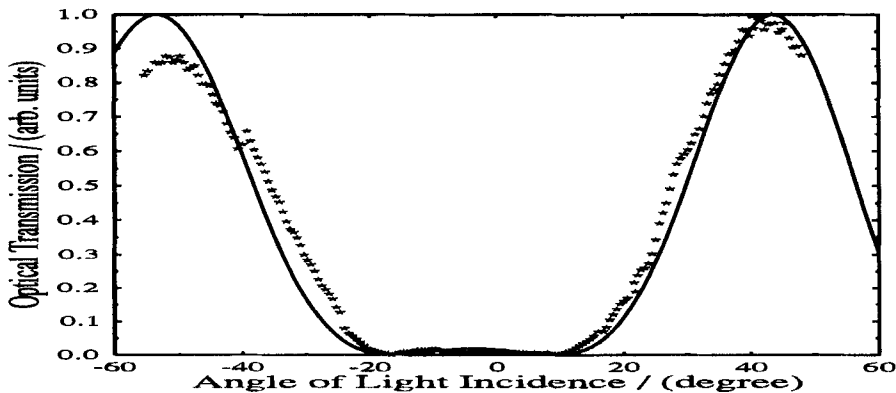


Figure 9: By means of the crystal rotation method, the average value of pretilt of the $19.6\mu m$ thick cell has been determined ($\theta_0 = 1.2^\circ$). The dots are the experimental data and the solid line is the theoretical fit.

several such images. Then, the influence of thermal fluctuations can be separated from the static pretilt. In the image depicted in Fig. 8a, after averaging over ten switching processes, we make out clearly a higher pretilt ($\approx 0.2^\circ$) on the left hand side than in the middle of the image. By means of the crystal rotation method, we have determined a value for the pretilt average over the cell plane which is $\theta_0 = 1.2^\circ$ (see Fig. 9). From our dynamical method, we have detected the same averaged value.

SUMMARY AND DISCUSSION

A novel method for the spatially resolved determination of thermal fluctuations and measurements of pretilt has been presented and applied to two types of cells with different surface preparations. No pretilt within the resolution of the experiment was found in cells with SiO_x 60° evaporation, and a pretilt of 1.2° was determined for antiparallel rubbed polyimide coating. It was shown that the local initial conditions of the director field caused by thermally activated fluctuations and surface pretilt influence the director dynamics and consequently the optical interference during a switching process. The transmission curves can be exploited for the determination of fluctuation amplitudes and pretilt. We obtain independent information by switching to different final voltages where only the switching time τ is influenced and the threshold voltage can be determined. An application of subcritical voltages prior to switching can be used to increase the effects of initial deviations of the director from planar orientation by destabilizing fluctuation modes. Furthermore, the relaxation time can be measured independently from the switching into the off state which yields relaxation times independent of the cell deformation. Thus, the only parameter in our fitting procedure is the initial director deformation $\theta_0(x, y)$.

The measurements are restricted to voltages near U_c . If the cells are switched to voltages much higher than U_c , transient patterns are observed¹² and higher deformation modes appear.

For larger pretilt angles, the method described in this article will be inexact because it assumes a sine director field in z direction. In cells with large pretilt, higher modes have to be considered in the switching mechanism from the beginning.

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