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Transient structures in the Fréedericksz transition

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An alternating electric field is applied to a planarly aligned nematic liquid crystal with a positive dielectric anisotropy sandwiched between two electrodes. This leads to a reorientation of the director when the critical threshold for the Fréedericksz transition is exceeded. Triggered by this reorientation process, patterns arise. In the experiments presented, a transient periodic structure showing up as a system of fairly regular stripes parallel to the initial orientation of the director is observed. This pattern seems to be caused by a secondary instability of the homogenous reorientation process.

The formation of transient patterns during the Fréedericksz transition in a nematic liquid crystal[1] has previously been studied both experimentally and theoretically.[2–6] The reorientation of the director in the applied field results in a definite elastic deformation in the sample. During the process of reorientation, hydrodynamics plays an important role since the director field is dynamically coupled to the velocity field, thus the rotation of the director induces a flow.[1] The striking feature of the reorientation process is the fact that it does not necessarily take place in a spatially homogeneous manner, but is rather accompanied by the formation of spatial structures. The physical reason for the patterns studied previously is, roughly speaking, that a transition with alternating parts of the director field turning clockwise and counterclockwise can have a faster response time when compared to the homogenous response.[2] As a result, a periodic pattern evolves. When the equilibrium director distribution according to the applied field is approached, the transient structures disappear.

According to the geometry used, the Fréedericksz transition can take place via a twist, splay, or bend deformation. Formation of transient patterns in the twist geometry has been studied intensively both theoretically[3] and experimentally.[4] A system of parallel lines was predicted and found perpendicular to the initial planar director alignment. Similar results have been obtained in lyotropic nematics.[5] The splay geometry was investigated in Refs. 2 and 6. Both *N*-(4′methoxybenzylidene)-4-(*n*-butyl)aniline (MBBA) and a lyotropic nematic were used and stripes oblique as well as perpendicular to the initial director alignment were found.

We use a planarly aligned thermotropic nematic with positive dielectric anisotropy and apply a sinusoidal electrical field to drive the splay Fréedericksz transition. Unlike the observations cited above, we observe a system of lines *parallel* to the initial orientation of the director, and we find no hint that the threshold for the formation of structures is significantly higher than the Fréedericksz threshold V_c. Also, the pattern does not disappear by an annealing process involving pairing up and annihilation of adjacent domain boundaries,[6] but the amplitude of the pattern decreases and vanishes locally.

We use 5CB (4-pentyl-4′-cyanobiphenyl),[7] a nematic liquid crystal with strong positive dielectric anisotropy ($\epsilon_\parallel - \epsilon_\perp \cong 12$). 5CB has a ratio of the elastic constants $K_2/K_1 = 0.615$, a typical value for low molecular weight nematics, which is well above 0.3, the critical value for the periodic splay-twist instability.[8] The sample is sandwiched between two parallel electrodes separated by a spacer of 100 μm. Planar alignment is achieved by rubbing the electrodes. The sample is thermostatted at $21.4 \mp 0.3\,^\circ$C. A harmonic electric field of frequency 70 Hz applied across the sample perpendicular to the initial director alignment induces the splay Fréedericksz transition. The data acquisition procedure for observing the transient patterns is the same as used for studies in electrohydrodynamic convection.[9] The sample is observed by means of a polarizing microscope with the polarization axis parallel to the director. Images of the pattern are recorded with a charge-coupled device (CCD) camera. The video signal is digitized with a resolution of 256 grey scales and a speed of 25 images per sec. The data acquisition and evaluation system described above allows us to detect and analyze scenarios with a duration of less than a second and thus we can use samples with a small thickness.

Figure 1 presents a measurement of the Fréedericksz threshold voltage V_c of our sample. Here we use the

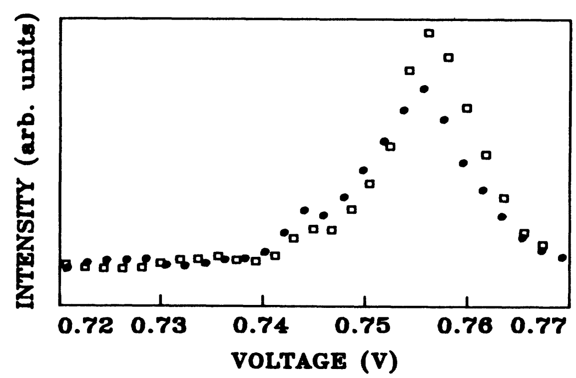

FIG. 1. Experimental determination of the threshold voltage V_c of the Fréedericksz transition. Open squares are obtained when increasing the voltage, solid circles during the decrease. Waiting time between two measurement points is 4 min.

birefringence technique which is the most sensitive one.[10] The sample is enclosed between crossed polarizers and the angle between the director and the axis of polarization is 45°. A change in the orientation of the director (the optical axis of the system) leads to variations of the He-Ne laser-light intensity passing through the sample. This occurs above the threshold value $V_c = 0.74$ V. The open squares correspond to measurements obtained by increasing the driving voltage and the solid circles are data recorded when stepping down the voltage. A waiting time of 4 min is used between consecutive voltage steps which is obviously not quite sufficient to make the difference between the two data sets vanish, but sufficient to pinpoint V_c on a percentage level. From here on a reduced voltage scale $\epsilon = (V - V_c)/V_c$ will be used.

Figure 2 shows the pattern formed during a ($\epsilon = 4.73$) reorientation process: white stripes are visible parallel to the initial director alignment. These lines are interpreted as caused by gradients of the director field during the transition. They are visible with high contrast when the light is polarized along the director, but hardly visible when the light is polarized perpendicular to this direction. Thus the stripes are caused by birefringence in the liquid crystal which leads to a strong refraction of the light at locations where the gradient of the director and thus the gradient of the refractive indices are large.[11]

We measure the light intensity along a line perpendicular to the initial orientation of the director (a horizontal line in Fig. 2). After increasing the voltage from a subcritical value (0.08 V rms, 70 Hz) to a supercritical one the intensity is measured every 0.08 sec. In Fig. 3 we show an example ($\epsilon = 4.73$) of the resulting curves plotted on top of each other with a time interval of 0.32 sec. Both the time necessary to build up the pattern and the transient nature of this structure is thus clearly demonstrated. Note that the intensity peaks, after reaching a maximum, fade away smoothly without changing their position. This means that the lines of the pattern cannot be interpreted as domain boundaries between two equivalent reorientations (with positive and negative splay angle) of the director. These kinds of defect lines are clearly distinguishable from the stripes of the structure; they are observable with any direction of the light polarization contrary to the pattern which is hardly visible by a polarization direction perpendicular to the lines. We saw disclinations at some parts of the sample, but we avoided them at the intensity measurements. Within the part of the cell observed here, the splay has the same sign, although its magnitude does not seem to be uniform during the transient.

In Fig. 4 we present the Fourier transform of the light-intensity curves given in Fig. 3. The aim of transforming the intensity lines is to get an idea about the wavelength of the transient pattern. The pattern, however, is not very regular at higher values of ϵ, thus no well-defined peak in the spectrum is observable. We therefore preferred to get a measure of the mean wavelength in the space domain by counting the intensity maxima along the line perpendicular to the director.

To obtain a value for the amplitude of the modulation, we measure the intensity along a line perpendicular to the director. From this we subtracted the intensity measured

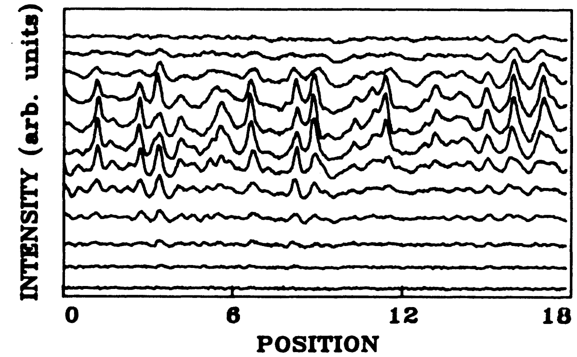

FIG. 3. Light intensity measured along a line perpendicular to the director alignment at different time intervals after applying the electric field. The lowest curve corresponds to $t = 0.32$ sec, the next ones follow within time intervals of 0.32 sec. At $t = 0$ the 70-Hz ac voltage is changed from 0.08 V to $\epsilon = 4.73$. The position is measured in units of the cell height.

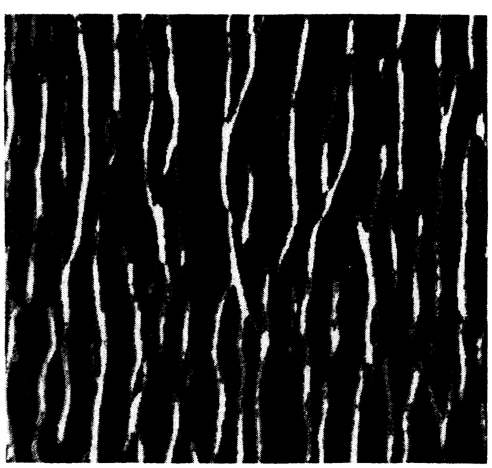

FIG. 2. The image (1.8×1.8 mm^2) of the transient pattern formed during the splay Fréedericksz transition in 5CB in polarized light. Stripes are parallel to the initial director alignment as well as to the direction of the light polarization. The applied electric field is perpendicular to the plane of the pattern.

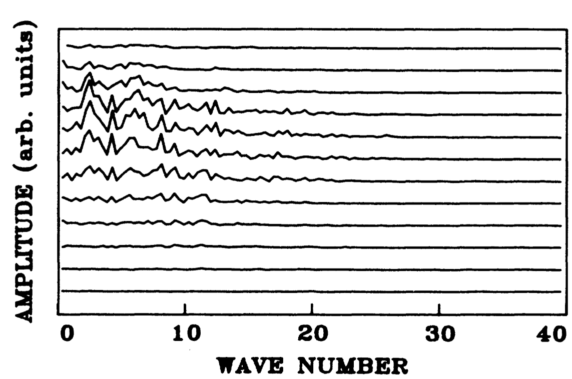

FIG. 4. Fourier transforms of the intensity lines given in Fig. 3. The wave number is scaled with respect to the height of the cell.

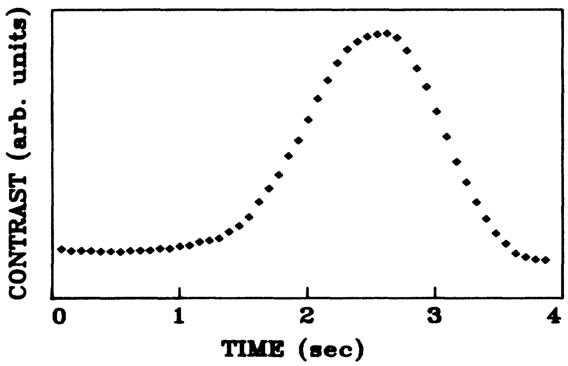

FIG. 5. Time dependence of the pattern contrast determined as the rms value of the light intensity measured along a line perpendicular to the director for the measurement shown in Fig. 3.

at a subcritical voltage and determined the quadratic deviation of this light intensity from its mean value. This rms value characterizes the contrast of the line. It is shown in Fig. 5 as a function of time for the measurement presented in Fig. 3.

From the experimental data presented in Figs. 3–5 several pieces of information can be extracted: the time t_m necessary for the pattern to build up (the location of the maximum in Fig. 5), the typical wavelength of the transient structure, and the maximum of the rms intensity. These three values are plotted for different voltage steps in Fig. 6. In the lower part the reciprocal value of the time t_m elapsed between the application of a supercritical voltage to the sample and the detection of the maximum contrast of the intensity is presented. The time t_m ranges from 0.7 sec for large voltage steps to about a minute for steps slightly exceeding the critical value. The maximum of the rms intensity is shown in the middle. The upper part of Fig. 6 shows the typical wave number of the transient patterns. The tendency to form patterns with a larger wave number at larger voltage steps is clearly demonstrated and very reminiscent of the data shown in Ref. 2.

From the middle part of Fig. 6 one can determine the threshold voltage above which the periodic structures appear by extrapolating to the value where the contrast is zero. Within the resolution of our data, there is no detectable difference between this value and the Fréedericksz threshold. This fact is contrary to the case of transient lines found in other systems perpendicular to the initial director alignment.[2,6]

In summary, we found an unexpected and unexplained pattern formation process. Unlike the experiments published before, we used an electric instead of a magnetic field to drive the transition. Although this is not expected to make a difference, we have checked the effect with a magnetic field in the same geometry and found the same direction of the stripes there. To rule out a conductivity effect, the experiments were carried out with different frequencies between 10 Hz and 10 kHz, which did not affect the pattern. To rule out an effect of the specific geometry of our sample, we performed measurements with cell

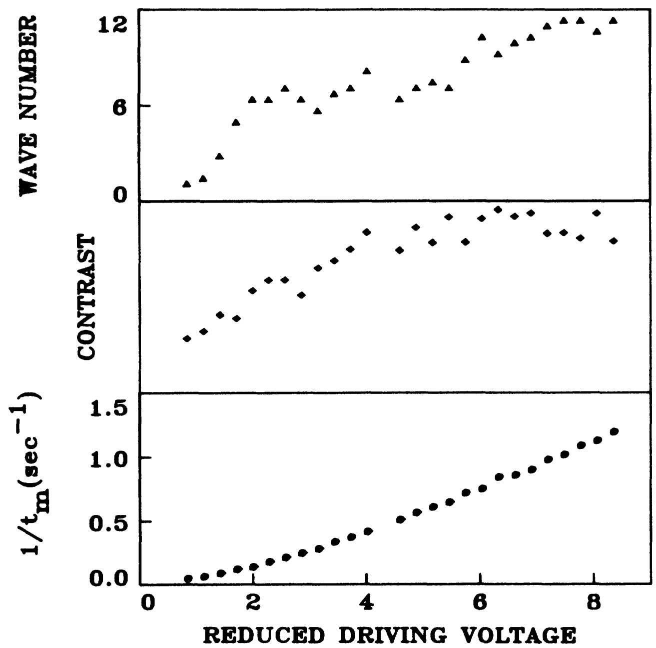

FIG. 6. The reciprocal value of the time for the pattern to reach its maximum amplitude as a function of the reduced voltage $(V-V_c)/V_c$ is shown in the lower part. The contrast reached after this time t_m is given in the middle. The upper part shows the mean wave number of the transient pattern.

thicknesses ranging from 15 to 250 μm, again showing no qualitative difference. Within the 250-μm cell, the patterns are clearly larger and the rise and decay times of the patterns are longer compared to the measurements with the 100-μm cell presented here, but the structures were *parallel* to the director in all the experiments.

Within the framework of the existing theories,[2–6] patterns normal or oblique to the director orientation would have been expected. These theories, however, are all based on a linear stability analysis of the unperturbed state. They check for the existence of a fastest growing mode with a wave number not equal to zero. If such a mode exists, however, it leads to a periodic splay field with alternating sign. Since the two directions are equivalent the domain boundaries constitute defect lines. Because we do not have this kind of structure, it seems to be more appropriate to assume a homogeneous reorientation as a starting condition and to check for a pattern-forming instability of this nonequilibrium state. A preliminary analysis with simplifying assumptions (artificial boundary conditions, large homogenous director distortion) indicates that such an instability is possible. A detailed analysis is in progress.

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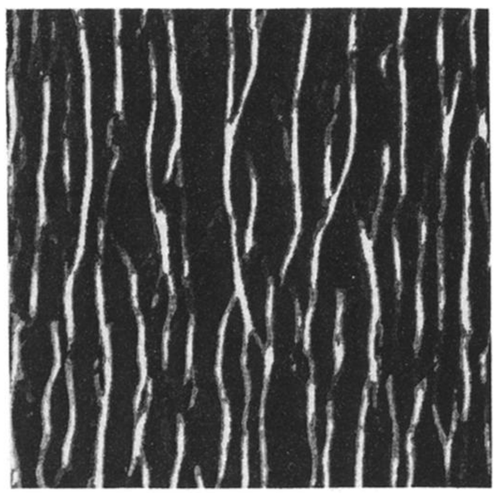

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