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Smectic C* layer directional instabilities in cells with twist geometry

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We have investigated the formation and structure of horizontal chevrons, as well as the reorientation dynamics of smectic layers under applied asymmetric electric fields in cells with a twist geometry. The tilted layer structure of horizontal chevron domains is found to be rotated by an angle approximately equal to the twist angle α, as compared with parallel rubbed substrates, $\alpha = 0°$. The time of horizontal chevron formation decreases slightly with increasing twist angle. The smectic layer reorientation under application of time-asymmetric electric fields is found to be enhanced for reorientation into the direction of twist, while it is hindered for reorientation out of the direction of layer twist. Increasing the twist angle results in a basically linear increase/decrease in the reorientation velocity, depending on field asymmetry direction. The electro-optic behaviour of twist cells with inclined smectic layers is outlined and compared with measurements performed on cells with monostable, parallel anchoring conditions.

1. Introduction

The formation of horizontal chevrons under application of symmetric electric fields, i.e. the occurrence of smectic C* domain structures with the smectic layer normal inclined at an angle with respect to the rubbing direction, often equal to the director tilt angle, has been demonstrated quite some time ago by Patel and Goodby [1]. Their formation process [2] and its dynamics [3] were investigated more recently. For parallel rubbed cells with planar, monostable anchoring conditions, these domain structures are formed by an irreversible texture transition from a vertical chevron or unoriented layer structure, either in the vicinity of a N*–SmC* [1] or SmA*–SmC* [2, 3] transition, under *symmetric* electric field conditions. For parallel rubbed substrates, the resultant structure is a domain texture with smectic layer orientations inclined to either side of the rubbing direction (figure 1), in equal distribution. The defect-mediating domains of opposite layer inclination (domain wall) are approximately several micrometers in width and of the vertical chevron type.

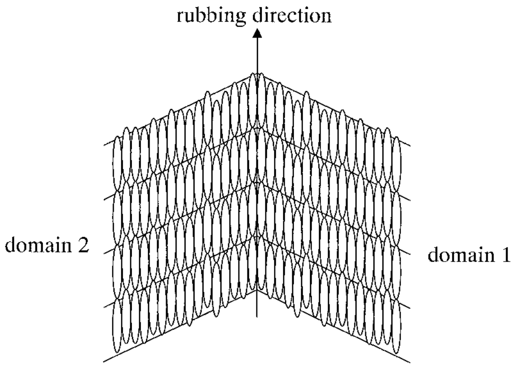

Figure 1. Schematic representation of the horizontal chevron domain structure, here without indication of the helical SmC* superstructure.

This smectic layer folding is a structure formation process, which is linked to the more general phenomenon of smectic layer directional instability under application of electric fields. In recent years, it has been shown that by application of an *asymmetric* electric field to a chiral smectic phase, a reversible reorientation of the smectic layer normal in the substrate plane can be observed. This has been demonstrated for the SmA* phase [4, 5],

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as well as the ferroelectric [6–8] and antiferroelectric [9, 10] modifications of the SmC* phase. Reversal of the electric field asymmetry, which can be either an amplitude or time asymmetric waveform (with or without a d.c. component [11, 12]), reverses the reorientation direction. The reorientation process is in fact not a rotation of whole domains into the opposite layer inclination direction, but rather a domain growth process of those domains favoured by the particular field asymmetry [13]. It is associated with permeation flow along the rubbing direction and subsequent domain wall motion [14]. The dynamics of this reorientation process, often also referred to as 'smectic layer rotation', is strongly dependent on externally applied conditions, such as electric field asymmetry ratio, amplitude, frequency or cell gap [12], as well as other parameters such as enantiomeric excess (magnitude of the spontaneous polarization and electroclinic coefficient) [15], smectic polymorphism (magnitude of the tilt angle) [16], ionic contamination [13] or substrate layer treatment [17, 18]. The smectic layer directional instability is a rather general phenomenon observed in chiral smectics under asymmetric electric fields and generally undesirable from the applicational point of view. It can be suppressed by the introduction of a texture stabilizing polymer network [19, 20], without dramatically changing the electro-optic performance (tilt angle, spontaneous polarization, response time) of the ferroelectric liquid crystal.

Here we would like to clarify in more detail the influence of the aligning substrates on the structure of horizontal chevrons and the dynamics of the smectic layer reorientation by investigation of cells exhibiting a twist geometry.

2. Experimental

The liquid crystal used in our studies is a commercially available epoxy compound, 4-[(*S*,*S*)-2,3-epoxyhexyloxy]-phenyl 4-decyloxybenzoate, from Aldrich. Its phase sequence on cooling is given by: I 95 N* 79.5 TGBA* 79.2 SmC* 54 SmI* 39 Cr (temperatures in °C). Twist cells were prepared such that the rubbing direction of the top substrate made an angle α with that of the bottom substrate. Angles were chosen to be $\alpha = -15°$ (corresponding to a counterclockwise, right-handed twist), $\alpha = 0°$ (corresponding to parallel alignment) and $\alpha = 5°, 10°, 15°, 20°, 25°$ and $30°$ (corresponding to clockwise, left-handed twists). Cells were prepared by spin coating ITO covered glass substrates with a polyimide solution at 3500 rpm for 30 s and tempering at 200°C for 3 h. These substrates were then unidirectionally rubbed by a rotating velvet cloth at the appropriate angles. Cells were assembled under slight pressure by dispersing spacer beads of diameter 6 µm within the UV curable glue, to assure a uniform cell gap.

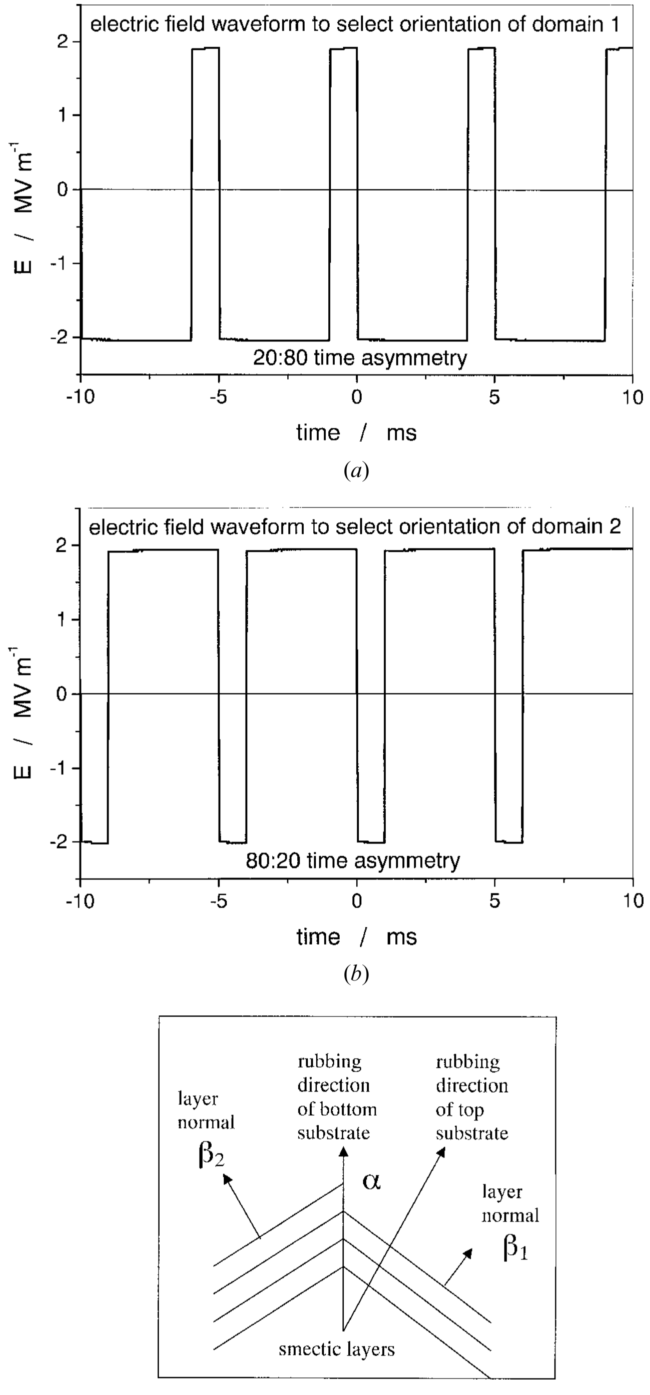


Figure 2. Applied electric waveform to select the orientation of (*a*) domain 1 and (*b*) domain 2; (*c*) indication of the nomenclature used, α = angle between the rubbing direction of the bottom and top substrate (positive for clockwise left-handed, and negative for counterclockwise right-handed twist), β_1 = angular in-plane position of the smectic layer normal of domain 1, β_2 = corresponding position of the domain 2 smectic layer normal.

Texture images were taken under crossed polarizer conditions by video recording and subsequent use of frame-grabber software (Adobe Premiere). The smectic layer reorientation was followed optically by polarizing microscopy (Leitz) during application of time-asymmetric electric fields produced by a Philips PM5138 function generator in combination with a F20A power amplifier (FLC Electronics). During the measurements the temperature was controlled within ±0.1 K by a Mettler FP80 temperature controller with FP82 hot stage. Electro-optic measurements were carried out by recording the transmission signal with a photodiode on a HP 54603B oscilloscope and data transfer to a PC. Experimental conditions were as follows: cell gap $d = 6\,\mu\mathrm{m}$, electric field amplitude $\mathbf{E} = 2\,\mathrm{MV\,m^{-1}}$, frequency $f = 200\,\mathrm{Hz}$, reduced temperature $T_\mathrm{C} - T = 1.5\,\mathrm{K}$ and varying time asymmetry (for example 20 means a time asymmetry ratio of 20% positive to 80% negative field, and other numbers correspondingly, 50:50 relating to symmetric field conditions). Figures 2(*a*) and 2(*b*) show the applied electric waveforms required to select the orientations of domains 1 and 2, respectively. For the electro-optic investigations, a symmetric electric field of frequency $f = 60\,\mathrm{Hz}$ was used, leaving other parameters unchanged. In figure 2(*c*) the notation of angles is schematically depicted: α denotes the twist angle, i.e. the angle between the rubbing direction of the bottom and top substrate plates. The direction of the polarizer was chosen such that it corresponds to the rubbing direction of the bottom substrate, set to $\phi = 0°$, β_1 and β_2 denote the

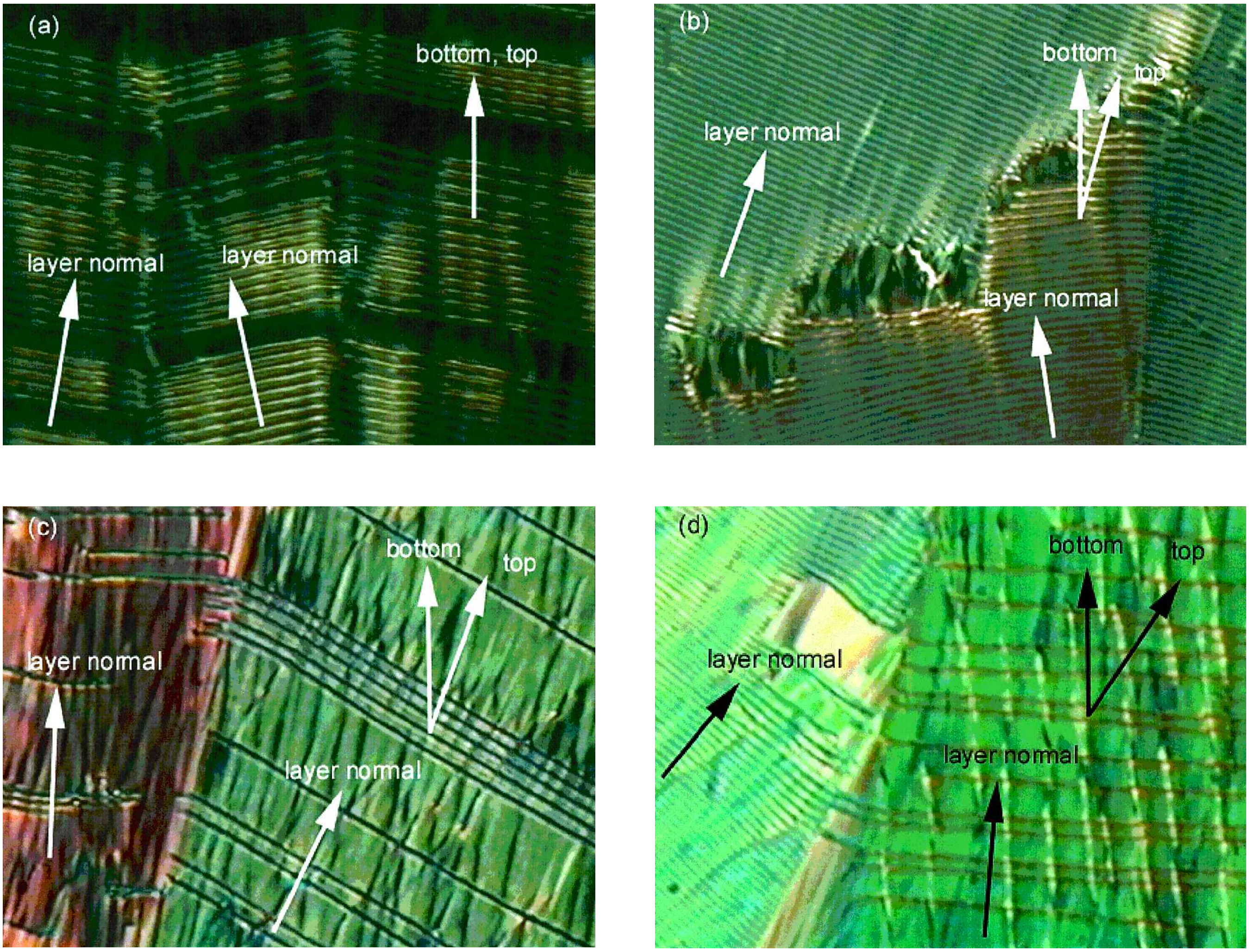


Figure 3. Texture photographs of the horizontal chevron domain structure for (*a*) $\alpha = 0°$, (*b*) $\alpha = 15°$, (*c*) $\alpha = 20°$ and (*d*) $\alpha = 30°$. The helix lines, although not fully developed due to the small cell gap, directly give the orientation of the smectic layer plane. It can be seen that the whole horizontal chevron domain structure is formed under an angle approximately equal to the twist angle α, as compared with that obtained for parallel boundary conditions. The shown image size is $180\,\mu\mathrm{m} \times 140\,\mu\mathrm{m}$ each.

angular ϕ direction of the smectic layer normal for clockwise and counterclockwise inclined domains, respectively (see also figure 1).

3. Experimental results and discussion

3.1. *Horizontal chevrons*

The effect of introducing a smectic layer twist, by non-parallel rubbing of the bottom and top substrates, on the structure of the horizontal chevron domain texture is shown in figure 3 for several twist angles, after induction of the horizontal chevron domain texture from an unoriented sample by a symmetric electric field. Figure 3 (*a*) shows the case of parallel rubbed substrates, $\alpha = 0°$. Smectic layers are inclined to either side of the rubbing direction by an equal angular value, generally equal to the director tilt angle θ. As the cell gap is only slightly larger than the SmC* pitch, the equidistant line pattern due to the helix formation is not fully developed. Nevertheless, the orientation of smectic layer planes can clearly be deduced, being parallel to the line pattern observed. In regions where the helix is not developed, it can clearly be seen that the direction of the molecular long axis, i.e. the director, points along the rubbing direction, leading to extinction in both domain types. Subjecting the liquid crystal to a twist geometry results in a clear change of the smectic layer arrangement, figures 3 (*b*)–(*d*). The frame of reference is now rotated by the amount α, having smectic layer normals of the two domains oriented at angular positions $\beta_1 = \theta + \alpha$ and $\beta_2 = -\theta + \alpha$, respectively. For rotation of the rubbing direction of the top substrate, horizontal chevron domains are also rotated in the same direction.

It should also be mentioned that the area distribution of domains 1 and 2 is no longer equal, as was observed for parallel rubbed substrates. One domain type, that with a smectic layer inclination in the direction of the layer twist, is clearly preferred (not shown in the texture photographs of figure 3), and this can be attributed to the fact that the monostable boundary conditions of the top substrate already favour the layer inclination in this direction. A quantitative evaluation of the respective textures is given in figure 4, depicting the in-plane angular positions β_1 and β_2 of the two domain types as a function of substrate twist angle α.

The introduction of a layer twist results in formation of the horizontal chevron domain structure rotated by an angle approximately equal to the twist angle, with respect to the structure observed for parallel rubbing, $\alpha = 0°$. In principle, both lines of the linear fit in figure 4 should be parallel, exhibiting a constant rotation of the horizontal chevron layer structure, and within the error limits of our investigations (cell orientation in the hot stage, hot stage orientation with respect to the rubbing direction of the bottom substrate), the above outlined behaviour is clearly observed. Figure 5 depicts the time of horizontal chevron formation from an unoriented smectic layer structure by application of a symmetric electric field. The formation time is found to decrease slightly with increasing twist angle α, which can be attributed to the fact that one direction of the boundary conditions already favours a layer inclined structure.

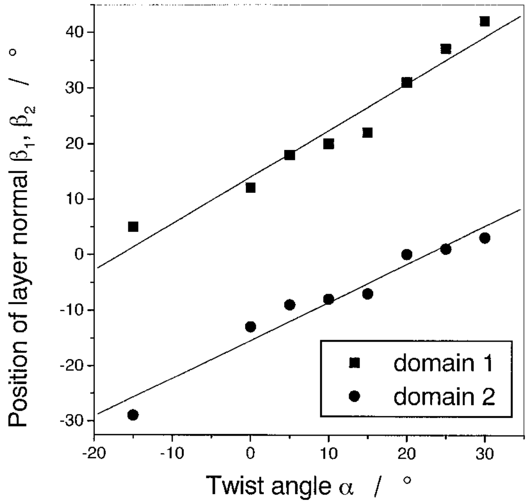


Figure 4. Angular in-plane orientations β_1 and β_2 of the smectic layer normals of the two domain types as a function of twist angle α.

3.2. *Smectic layer reorientation*

The reorientation of smectic layers, i.e. the smectic layer directional instability under application of asymmetric electric fields, is demonstrated by the texture photographs of figure 6 for a cell with parallel boundary conditions, $\alpha = 0°$, and the rubbing direction oriented along one of the polarizers. Starting from an unoriented

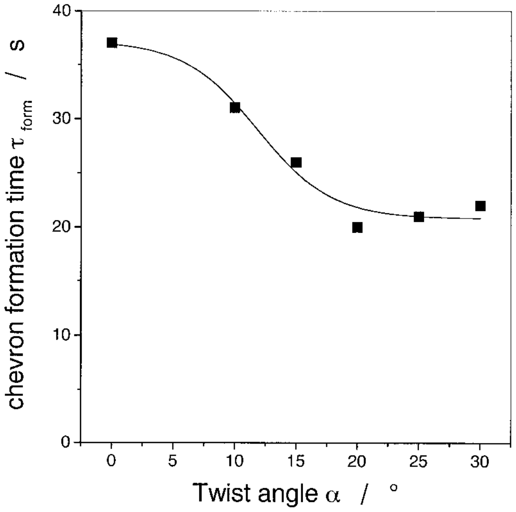


Figure 5. Time of horizontal chevron formation from an unoriented layer structure as a function of twist angle α.

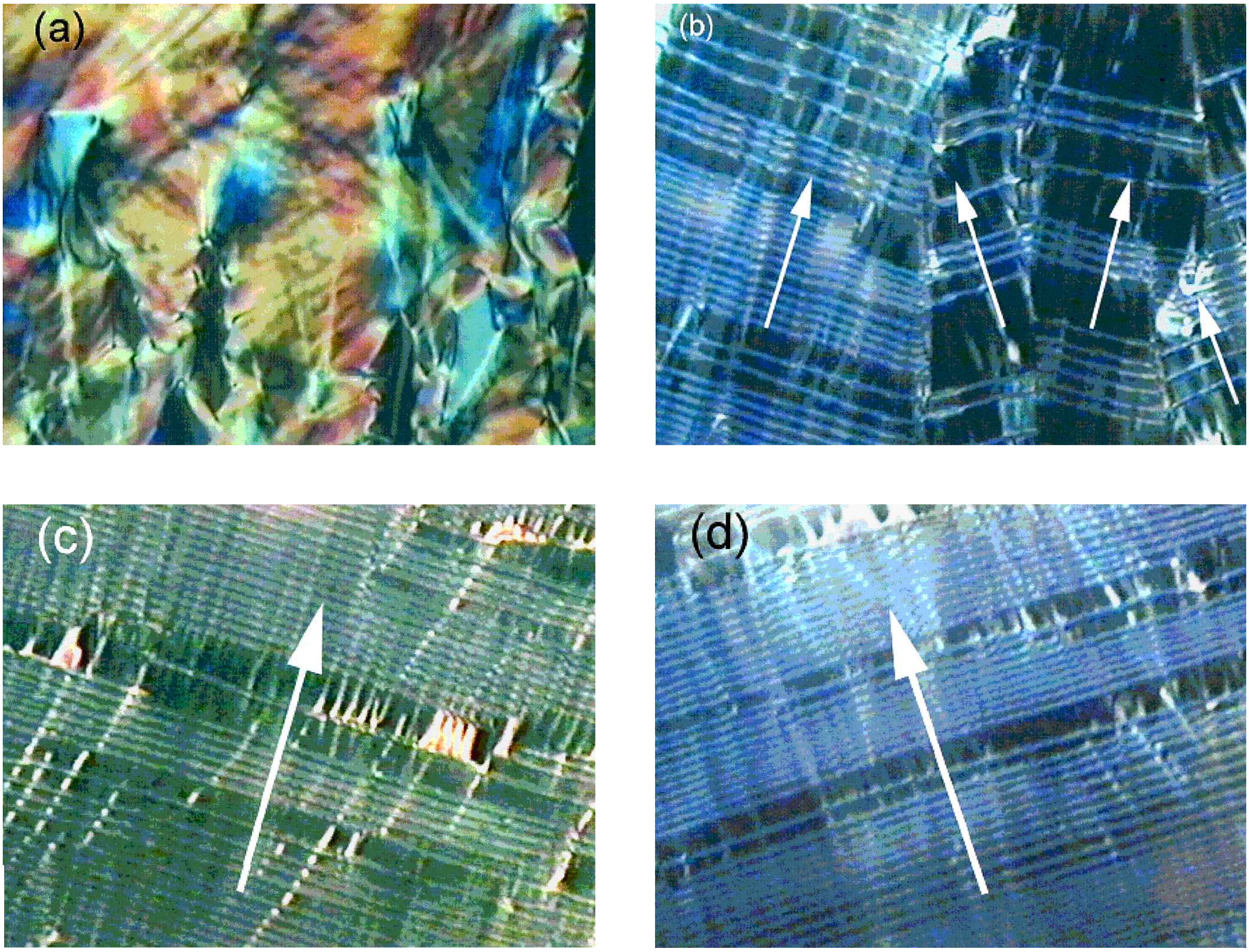
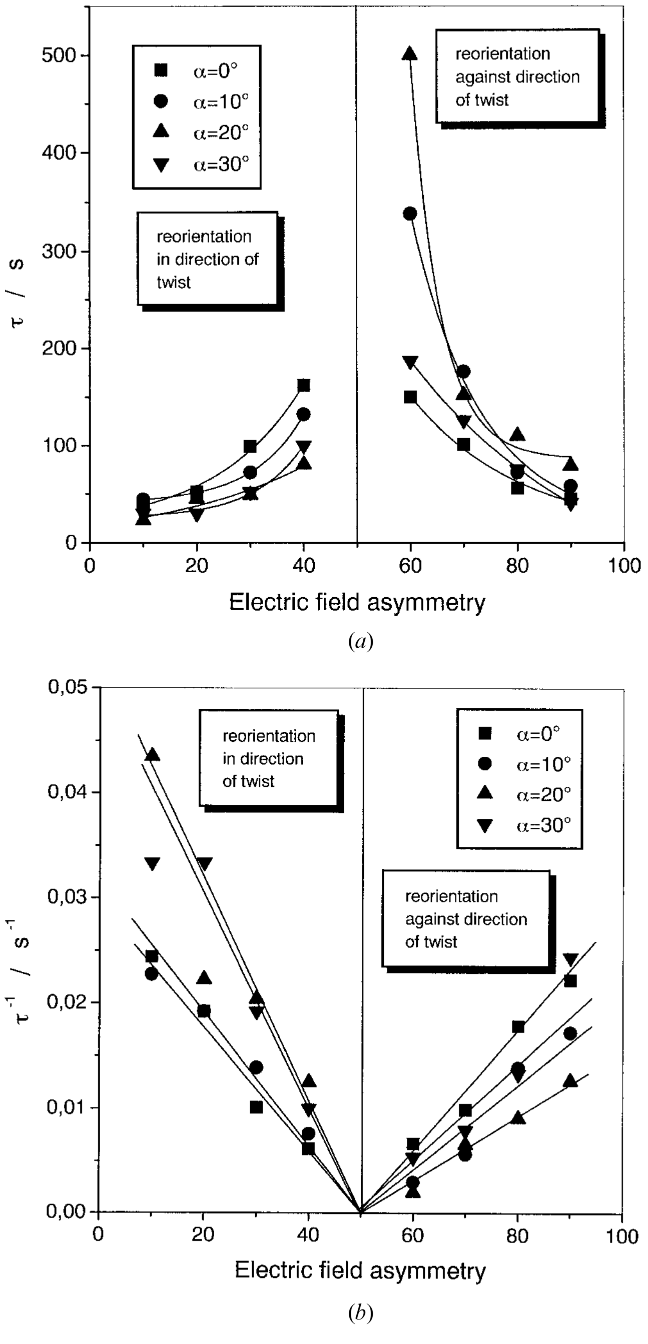


Figure 6. Texture photographs demonstrating the reorientation of smectic layers for the cell with parallel anchoring conditions, $\alpha = 0°$ (helix lines correspond to the smectic layer plane). (*a*) Unoriented texture after transition into the SmC* phase; (*b*) horizontal chevron domain texture after application of a symmetric electric fields; (*c*) reoriented layer structure after application of a 20:80 time asymmetric electric field; (*d*) smectic layer structure after reversal of the field asymmetry to 80:20. The shown image size is 290 µm × 220 µm each.

sample, figure 6 (*a*), obtained by merely cooling from the cholesteric (N*) into the smectic C* phase, a symmetric electric square wave field is applied to induce the horizontal chevron domain structure, figure 6 (*b*)—only parts of the two domains, including the domain wall, are shown. Helix lines again correspond to the smectic layer planes. In areas where the helix is not fully developed, we can observe from the black state that in both domain types the director is oriented along one of the polarizer directions, i.e. along the rubbing direction. After application of a 20:80 time asymmetric square wave electric field, domain type 1 has grown over the entire electrode area, exhibiting a sample with uniform smectic layer inclination, rotated clockwise with respect to the rubbing direction, figure 6 (*c*). Reversal of the electric field asymmetry to 80:20 results in a layer reorientation process via domain nucleation and growth until a structure with smectic layers inclined in the opposite direction is observed across the whole electrode area, figure 6 (*d*). The dynamics of this smectic layer reorientation process are now investigated for cells with the twist geometry, $\alpha \neq 0°$.

In accordance with earlier investigations [12], the time of reorientation of one domain type into the other increases strongly with decreasing electric field asymmetry; thus on approaching symmetric field conditions (50:50) it diverges, figure 7 (*a*). Accordingly, the inverse reorientation time, being a measure of the reorientation



(*a*)

(*b*)

Figure 7. (*a*) Smectic layer reorientation time τ as a function of field asymmetry for several twist angles α; (*b*) corresponding inverse reorientation time τ^{-1}, as a measure for the layer reorientation velocity.

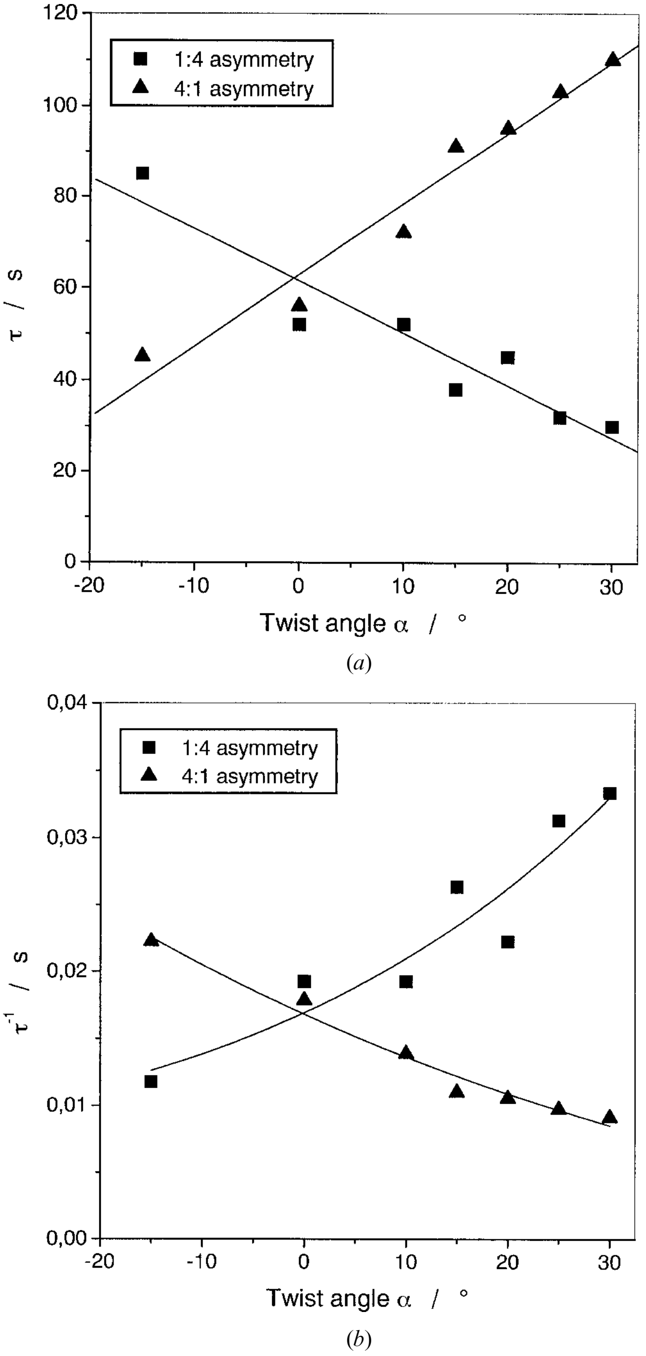
velocity, decreases with decreasing field asymmetry and vanishes for symmetric field conditions, figure 7 (*b*). For the twist geometry, we observe a pronounced difference in reorientation times whether the reorientation is performed into the direction of the twist or out of it. Reorientation processes in the direction of twist (asymmetry ratios < 1) are clearly faster than those out of the twist direction (asymmetry ratios > 1). For reorientation into the twist direction $\alpha \neq 0°$, the process is also *slowest* for parallel rubbed substrates ($\alpha = 0°$, squares in figure 7), while for the reorientation process out of the twist direction, it is *fastest* for $\alpha = 0°$. Note also, that for parallel rubbed substrates the reorientation times or respective velocities are symmetric around the 50 : 50 field condition, while this is not the case for the twist geometry.

In figure 8 the behaviour is depicted for a different measurement series at 20 : 80 and 80 : 20 time asymmetry ratio of the applied electric square wave field as a function of the twist angle α. Reorientation times of smectic layers into the twist direction (squares) decrease with increasing twist angle ($\alpha > 0°$), while they increase for the reverse process (triangles), see figure 8 (*a*). For parallel rubbed cells, the times are practically equal for 20 : 80 and 80 : 20 asymmetry, as expected. The respective behaviour for the reorientation velocity is depicted in part (*b*) of figure 8. A cross-experiment for cells with twist angle $\alpha = -15°$ reveals that here the opposite reorientation dynamics are observed: the reorientation time is faster for 80 : 20 than for 20 : 80 time asymmetry. In this case, the reorientation velocity for the counterclockwise rotation is larger than for the clockwise rotation. In comparison with the cell with $\alpha = +15°$, reorientation times and velocities are approximately equal, but at reversed field asymmetry.

Different preferred alignment directions of the bottom and top substrates introduce a mechanical twist deformation of smectic layers. For the reorientation process, this acts like an additional electric bias field in the appropriate direction (in this case $\mathbf{E}_{\mathrm{bias}} < 0$ for $\alpha > 0°$). The dynamic behaviour of the reorientation process in twist cells is somewhat similar to the effect of introducing a polymer network, formed at a layer configuration, which has already been rotated in one direction [20] (in this case, clockwise), thus also introducing an effective elastic field. Nevertheless, there is a distinct difference. For the network-stabilized materials, the reorientation process in general became slower as compared with the non-stabilized sample, while for the twist cell geometry it can be observed to be faster than for the non-twisted geometry.

3.3. *Electro-optic measurements*

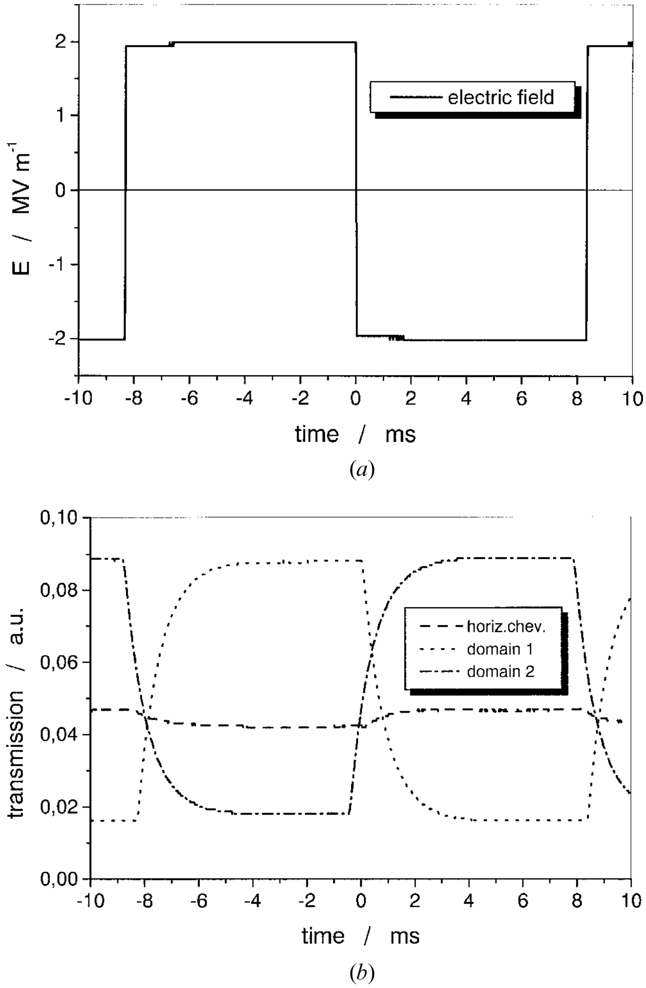
The electro-optic behaviour of a cell with parallel boundary conditions, $\alpha = 0°$, is depicted in figure 9. In figure 9 (*b*) the electro-optic response to a symmetric square wave field of amplitude $\mathbf{E} = 2\,\mathrm{MV\,m^{-1}}$ and frequency $f = 60\,\mathrm{Hz}$, shown in figure 9 (*a*), is given for the different geometries discussed. The polarizer direction is chosen along the rubbing direction. For an absolutely



(*a*)

(*b*)

Figure 8. (*a*) Smectic layer reorientation time τ at 20:80 and 80:20 field asymmetry as a function of the twist angle α; (*b*) corresponding inverse reorientation time τ^{-1}, as a measure of the smectic layer reorientation velocity.



(*a*)

(*b*)

Figure 9. (*a*) Applied electric field during the transmission measurements. (*b*) Electro-optic response of a cell with parallel boundary conditions for different smectic layer configurations. Opposite layer inclination with respect to the rubbing direction results in a complementary electro-optic response.

equal distribution of horizontal chevron domains, one would expect a constant transmission without a dark position because, as the director of one domain type is switching along the tilt cone 2θ into the direction of the polarizer, the director of the other domain reorients out of this direction by the same angle. This behaviour is confirmed by the dashed line of figure 9 (*b*), although a small modulation of the transmitted light intensity is observed; this is due to a slightly non-equivalent domain area distribution within the field of view of the microscope. The dotted line of figure 9 (*b*) depicts the transmission response to a symmetric square wave field after application of a 20:80 time asymmetric field for a suitable time period, selecting domain 1 across the whole electrode area. The transmission now changes from a dark state (director along one polarizer, negative field) to a bright state (positive field). This electro-optic response is reversed when we subject domain 2 (obtained after applying an 80:20 asymmetric field) to the same symmetric switching field, see figure 9 (*b*), dash-dotted line. The outlined electro-optic behaviour is in accordance with the structure of the horizontal chevrons and the layer-rotated configurations.

Similar measurements for two twist cells, one with a clockwise twist of $\alpha = +15^\circ$ and the other with a counter-clockwise twist of $\alpha = -15^\circ$, are shown in figure 10; electric field as in figure 9 (*a*). Again, one polarizer is oriented along the bottom rubbing direction. As the frame of reference is now rotated by $\pm\alpha$, compared with parallel boundary conditions, the electro-optic responses of domains 1 and 2 of each cell are not complementary, as can be seen for curves (*a*) (solid) and (*b*) (dashed) for a cell with $\alpha = -15^\circ$, as well as for curves (*c*) (dotted) and (*d*) (dash-dotted) for a cell with $\alpha = +5^\circ$. Complementary electro-optic behaviour is now observed for domain 2 of a cell with $\alpha = -15^\circ$ (selected by 80:20 asymmetry, curve (*a*)) and domain 1 of a cell with $\alpha = +15^\circ$ (selected by 20:80 asymmetry, curve (*d*)), as well as domain 1 of the cell with $\alpha = -15^\circ$ (selected by 20:80 asymmetry, curve (*b*)) and domain 2 of the cell with $\alpha = +15^\circ$ (selected by 80:20 asymmetry, curve (*c*)). A similar behaviour would be observed for any other pair of twist cells with $\pm\alpha$.

4. Conclusions

The introduction of a smectic layer twist by choosing different rubbing directions for the bottom and top substrates has a pronounced effect on the smectic layer directional instability. Horizontal chevron domain structures as a whole are found to be formed at an angle approximately equal to the twist angle, as compared with the cells with parallel boundary conditions. Their formation dynamics is slightly enhanced for increasing layer twist.

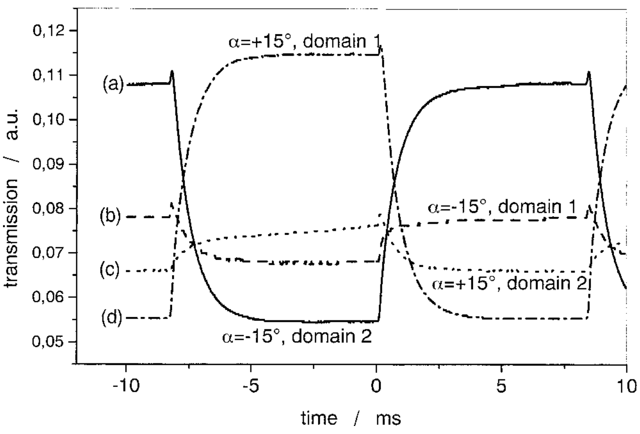


Figure 10. Electro-optic response of two twist cells with opposite twist direction, $\alpha = \pm 15^\circ$. A complementary electro-optic response is observed for opposite layer inclination (obtained for reversed field asymmetry) in cells of opposite twist sense. Electric field as in figure 9 (*a*).

Considering the dynamics of the smectic layer reorientation under asymmetric electric field conditions, reorientation times are observed to decrease for 'rotation' into the twist direction, while they increase for the reversed process. For reorientation into the twist direction, the reorientation velocity is larger than for parallel rubbed cells, while for the reverse process (reorientation out of the twist direction) it is slower. Generally, enhanced (hindered) reorientation dynamics is observed for 'rotation' into (out of) the twist direction, with a basically linear decrease (increase) in reorientation time as a function of increasing twist angle. The effective elastic twist field, introduced by non-parallel rubbed substrates, acts like an additional electric bias field.

For cells with parallel boundary conditions, the electro-optic response of layer configurations inclined to opposite sides of the rubbing direction (obtained by reversing the field asymmetry), is complementary. For twist cells, complementary electro-optic behaviour is observed for opposite layer inclinations (obtained for reversed field asymmetry), at opposite twist sense.

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