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Formation characteristics of horizontal chevron structures in ferroelectric liquid crystal cells

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The formation process of field induced smectic C* layer structures known as 'horizontal chevrons' has been investigated as a function of external parameters. Characteristic dimensions of field induced domains, as well as the dynamics of their formation process are reported for a variation of cell gap, applied electric field amplitude, frequency and temperature. The experimental data are discussed in terms of liquid crystal–surface interactions, director switching and effects of ionic motion.

1. Introduction

Ferroelectric smectic C* (SmC*) liquid crystals may exhibit a variety of director and layer configurations when prepared between two glass plates covered with an aligning layer, often rubbed polyimide. Generally, a chiral smectic C* phase exhibits a helical superstructure which is observable by an equidistant line pattern with the twist axis perpendicular and the lines parallel to the smectic layer plane [1, 2]. If the cell gap is reduced to the order of magnitude of or smaller than the helical pitch at planar boundary conditions, the helix is unwound by liquid crystal–surface interaction [3] and a bookshelf smectic layer configuration may be formed, with smectic layers perpendicular to the substrates [figure 1(a)]. The molecules are tilted with respect to the smectic layer normal. More often structures are observed where the smectic layers are tilted with respect to the substrate normal. The tilted layer configuration [figure 1(b)] is generally obtained for anti-parallel rubbed alignment layers, while the vertical chevron structure is formed for parallel rubbed substrates [figure 1(c)]. This smectic layer configuration has been discussed previously [4, 5].

The transition zones between two areas of vertical chevrons with opposite sign are called zigzag defects [6, 7]. They are of the quasi-bookshelf type. The application of an electric field to vertical chevron samples exerts a torque on the smectic layer, as the spontaneous polarization P_s is not parallel to the electric field vector. This may result in reversible or irreversible straightening-up of the smectic layers, as demonstrated by several authors [8–13]. The process can result in the

formation of smectic layer domain structures which were first reported by Patel *et al.* [14, 15] and resemble vertical chevron structures turned through 90° —so-called horizontal chevrons. It has been shown that the field induced domain formation can be regarded as a layer reassembly of vertical chevrons to the horizontal type

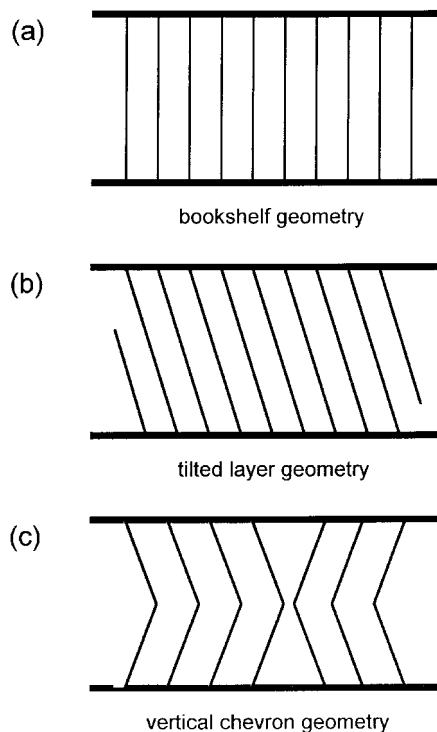


Figure 1. Schematic illustration of different smectic layer structures of SSFLC cells: (a) ideal bookshelf geometry, (b) tilted layer structure and (c) vertical chevron configuration.

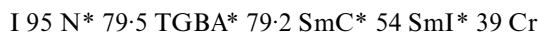
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[16]. During this process defects of the bookshelf type, mediating regions of opposite vertical chevron sign, are transformed into vertical chevron defects, mediating bookshelf type regions of opposite horizontal chevron structures [16]. The horizontal chevron structure is depicted in figure 2 as a top view of the cell for a helical sample, to indicate the smectic layer directions. The helix lines are parallel to the smectic layers. Two domain types of opposite smectic layer tilt with respect to the rubbing direction can clearly be distinguished. The borders or walls (bright lines) between domains of opposite layer tilt are vertical chevron defects of opposite sign. In regions where the SmC* helix is not fully developed, it can be seen that molecules are oriented along the rubbing direction, which coincides with the direction of one of the crossed polarizers. Thus these regions appear dark in both types of domains, even though the vectors of the spontaneous polarization are anti-parallel.

This behaviour is more clearly demonstrated in figure 3, while applying a bias voltage and thus unwinding the helix. With positive bias a certain domain type appears dark, while the molecules of the other domain type are oriented at an angle 2θ (twice the tilt angle) to the rubbing direction and appear bright [figure 3(a)] [16, 17]. With no bias field the molecules of both domain types are oriented along the rubbing direction, thus resembling a well oriented bookshelf texture. Regions between the two domain types are still visible as defect lines of the vertical chevron type [figure 3(b)]. Reversal of the bias field direction as opposed to figure 3(a), results in the inverse image [figure 3(c)]. The field induced domains are elongated along the rubbing direction, but there is no direct correlation between domain width and LC cell gap, as is observed for the so called 'striped texture' [18, 19].

2. Experimental

The liquid crystal used in this study was a commercially available compound, 4-[(S,S)-2,3-epoxyhexyloxy]phenyl 4-decyloxybenzoate (Aldrich Chemicals). Its phase sequence† on cooling is given by



The hexatic SmI* phase can be strongly supercooled. Phase transition temperatures did not significantly change for preparations in cells of different cell gap. Cells used

† A TGB phase has not been mentioned in the literature relating to this compound. However, our texture studies appear to reveal the presence of a very narrow TGB phase which seems to be TGBA*, although a TGBC* phase cannot be absolutely excluded. The nature of this phase is not however of relevance to the investigations discussed in this publication.

were commercially available (E.H.C., Tokyo, Japan) at various cell gaps between 2 and 15 μm with parallel rubbed polyimide orientation layers and active electrode areas of $4 \times 4\ \text{mm}^2$.

Texture analysis was performed with a Nikon OPTIPHOT2-POL polarizing microscope, equipped with a Sony Hyper HAD model SSC-DC38P digital video camera for direct computer image processing and storage. Image analysis was performed with software from Bergström Instruments AB. Average domain width values W_{av} were determined by averaging 40 measurements for each texture observation after inducing the domains. This involves reading off the domain width with respect to a calibration scale and measurements were confirmed for different cells. A statistical analysis was not carried out to determine the standard deviation, as trends, not absolute values, are under discussion and the statistics of domain width are not of real relevance to the dynamics of horizontal domain formation. The sample temperature was controlled with an Instec RTC-1 hot stage. Electric fields were applied to the samples with an OR-X model 410 function generator in combination with a high voltage amplifier model F400D from FLC Electronics. Electro-optic investigations were monitored by a Tektronix TDS 540 digital storage oscilloscope.

Cell gap dependent measurements were carried out with a symmetric 200 Hz square wave field of amplitude $1\ \text{MV m}^{-1}$ at 0.5 K below the SmC*-TGBA* transition ($T_{AC} - T = 0.5\ \text{K}$). The voltage dependence of the domain induction process was investigated for a $10\ \mu\text{m}$ cell, again for an applied electric square wave field of frequency $f = 200\ \text{Hz}$ at $T_{AC} - T = 0.5\ \text{K}$. Frequency dependent measurements were performed at respective parameters and applied electric field amplitude of $E = 1\ \text{MV m}^{-1}$. The temperature dependence of the domain formation process was investigated for analogous parameters. Investigations of the domain width distribution were carried out for $10\ \mu\text{m}$ samples subjected to a 200 Hz symmetric square wave field of amplitude $1\ \text{MV m}^{-1}$ at $T_{AC} - T = 0.5\ \text{K}$. Values were determined by counting the number of domain widths observed in an interval of $25\ \mu\text{m}$ for 400 measurements on 10 independently induced textures of the same cell. For each single measurement of the various series of the investigation, the samples were heated well into the cholesteric phase, then slowly cooled across the respective transitions into the ferroelectric SmC* phase to the temperature of investigation.

3. Results and discussion

One of the obvious questions in the investigation of field induced layer structures is that of the surface

memory effect. Do, for a given sample, horizontal chevron domains form at the same region of a sample, even after heating into the cholesteric phase and cooling back to the SmC* phase? This question can clearly be denied, as shown by the micrographs of figure 4(a)–(d), taken for the same sample at an identical position after heating into the cholesteric phase, successive cooling into the SmC* phase to $T_{AC} - T = 0.5$ K and inducing the domain structure by application of a symmetric square wave electric field of frequency $f = 200$ Hz, amplitude 1 MV m^{-1} for a $10 \mu\text{m}$ cell. The two different domain types form at arbitrary regions of the electrode area and are often elongated along the rubbing direction. Hence, in the formation process of horizontal chevron domain structures there is no surface memory effect involved.

The distribution of the domain width, as a characteristic dimension of the field induced domain formation of horizontal chevrons, is depicted in figure 5 for a $10 \mu\text{m}$ sample. Each measurement point represents the normalized counts of the domain width W within a specific interval of $25 \mu\text{m}$. The distribution shows a small domain width cut-off. The fact that we here deal with a domain width distribution represents a clear distinction from the striped texture, for which a delta function-like behaviour is observed (approximately constant stripe width, equal to the cell gap [19]).

The dependence of the average domain width W_{av} and the time of formation τ_{form} from the original texture, as a function of external parameters, is depicted in figures 6–9 for a variation of cell gap, electric field amplitude, frequency and temperature, respectively. The general process of the horizontal chevron domain formation was reported in ref. [16] as an irreversible layer straightening, preserving the molecular tilt angle with respect to the layer normal. It is observed that the average domain width increases with increasing cell gap. The time of formation shows an increase between $6 \mu\text{m}$ and $10 \mu\text{m}$ before saturation is reached (figure 6). For the field induced formation of horizontal chevrons, a material flow is necessary, as the formation process involves some mass transport. This material flow can be provided by convection due to ionic motion as well as the ordinary director switching. As the cell gap is increased, more liquid crystal material can behave in a bulk-like way, being less dominated by the substrate, allowing for easier material flow and thus leading to larger domains. As the formation process of horizontal chevrons is a domain nucleation and growth process, faster molecular rearrangement leads to larger domain sizes.

An increase in the applied electric field amplitude results in decreasing time of domain formation, while

the domain size increases (figure 7). For the layer rearrangement a mass transport, as can be provided by convection, is necessary. This is most easily achieved for high electric field amplitudes. Also the layer straightening is more pronounced by far for larger field amplitudes [9, 11, 12]. For the smectic layer reorientation the usual director switching is important. Also the induced tilt due to the electroclinic effect increases with electric field strength, leading to an increased molecular motion and thus encouraging the formation of larger domains. This is especially the case near phase transitions, here the SmC* to TGBA* transition at $T_{AC} - T = 0.5$ K.

For increasing frequency, an increase in the time of domain formation is observed, while the domain width is basically constant for low frequencies, before a strong decrease for frequencies above about $f = 500$ Hz can be detected (figure 8). We presume that the increase in domain formation time with increasing frequency is due to a combination of two effects. First, the material flow due to convection should decrease as the frequency increases, thus hindering the domain formation. At the same time, it should be increasingly difficult for the layer straightening to follow the increasing frequency. Both effects should lead to an increase in domain formation time. The decrease of domain width for high frequencies is caused by a vanishing saturated director switching, as can be confirmed by electro-optic investigations. The frequency of $f = 500$ Hz for which a starting decrease in domain size is observed, coincides with that where saturated ferroelectric switching ceases.

Lowering the temperature results in a decrease of the average domain size and an increase in formation time of the horizontal chevron domain structure (figure 9). Very close to the SmC* to TGBA* transition the liquid crystal material is soft. A strong decrease of the effective viscosity is generally observed in the vicinity (1–2 K) of the transition, approaching the cholesteric phase. This may account for the temperature behaviour discussed above, especially for the strongly changing values between 1 and 2 K below the transition.

4. Conclusions

Horizontal chevron structures can be formed in ferroelectric liquid crystal cells by the application of electric fields. Domains of opposite layer tilt with respect to the rubbing direction are formed. The layer tilt angle is equal to the director tilt angle [16], which means that the molecules at zero field lock into the direction of rubbing. The induction process of these domains is not related to a surface memory effect. Even though domains are often elongated along the rubbing direction, there is no correlation between domain width and cell gap. In contrast to the stripe-texture, we observed a domain

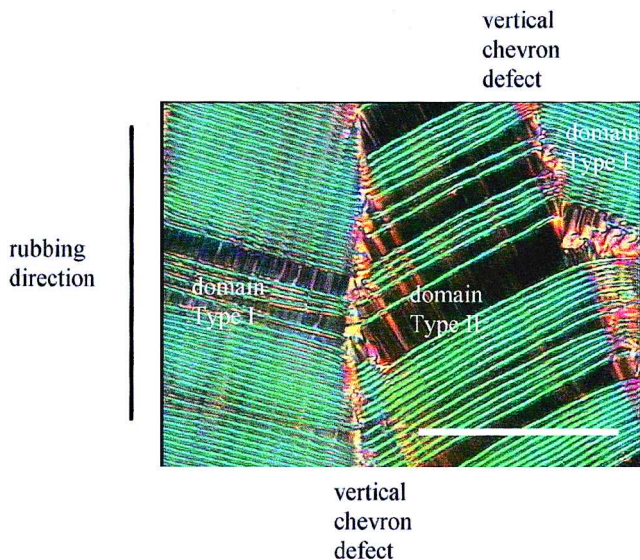


Figure 2. Texture micrograph of the horizontal chevron domain structure. The smectic layers of different domains are tilted in opposite directions with respect to the rubbing direction. This is indicated by the line pattern due to the SmC* helix, which is observed along the smectic layers. The bar is equal to 100 μm .

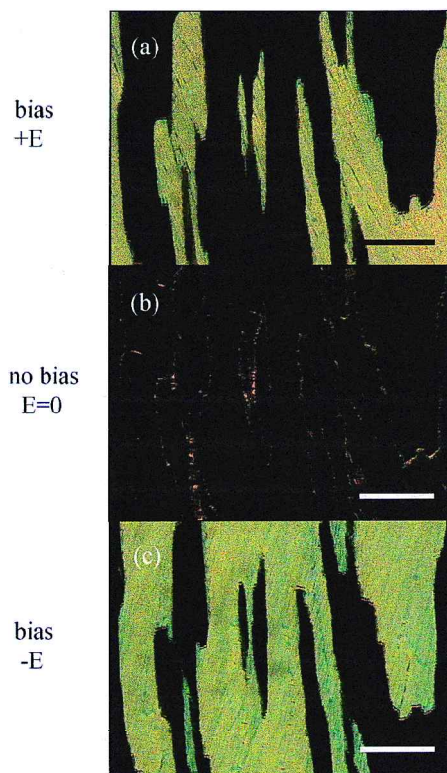


Figure 3. Horizontal chevron domain structure for (a) applied positive bias field, (b) zero field and (c) negative bias field. The micrographs clearly demonstrate a smectic layer and director configuration with molecules orientated parallel to the rubbing direction, and domains of opposite layer tilt and direction of the spontaneous polarization. The bar is equal to 200 μm .

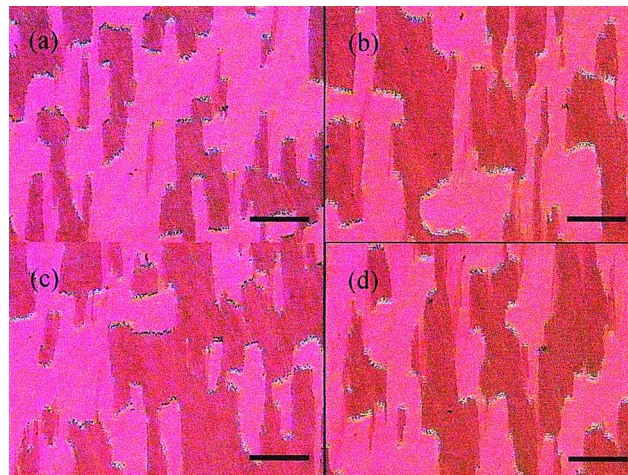


Figure 4. Texture photographs of horizontal chevron domains taken at the same region of the sample for several cycles of heating into the cholesteric phase, successive cooling into SmC* and application of an electric field to induce the horizontal chevron domain structure: (a)–(d) demonstrate that the horizontal chevron formation process is not a surface memory effect.

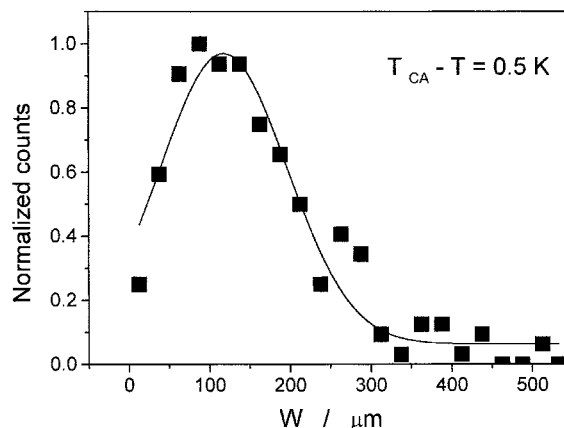


Figure 5. Domain width W distribution of field induced horizontal chevron domains. The data can be modelled by a distribution function with small domain width cut-off. Experimental conditions: 10 μm sample, 200 Hz symmetric square wave field, $E = 1 \text{ MV m}^{-1}$, $T_{\text{AC}} - T = 0.5 \text{ K}$.

width distribution instead of equidistance spaced lines of width equal to the cell gap [18, 19]. To obtain horizontal chevrons monostable anchoring (unidirectional rubbing) is needed.

For the domain formation of horizontal chevron structures a material flow within the material is necessary, which is generally provided by the director switching and convection due to ionic motion. The domain formation process is favoured by large cell gaps, high electric field amplitudes, low frequencies and

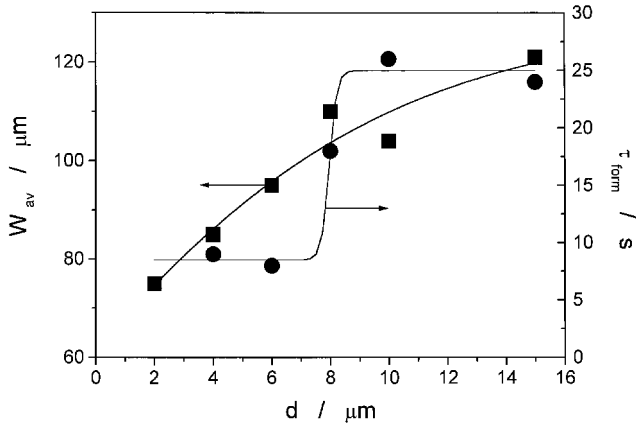


Figure 6. Cell gap dependence of the average domain width W_{av} (squares) and time of domain formation τ_{0rm} (circles). Experimental conditions: symmetric 200 Hz square wave field, $E = 1 \text{ MV m}^{-1}$, $T_{AC} - T = 0.5 \text{ K}$.

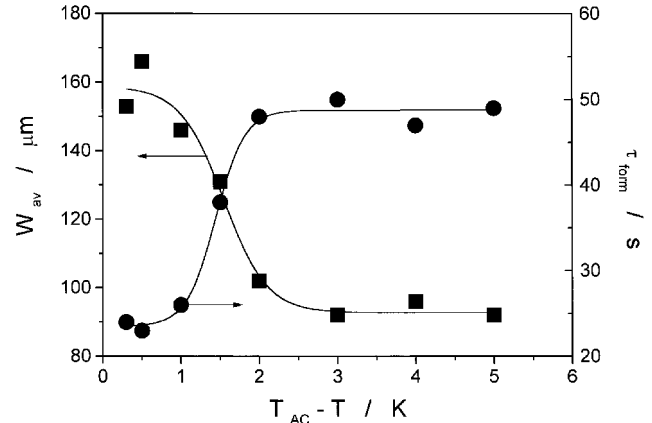


Figure 9. Temperature dependence of the average domain width W_{av} (squares) and time of domain formation τ_{0rm} (circles). Experimental conditions: $10 \mu\text{m}$ cell, 200 Hz square wave field, $E = 1 \text{ MV m}^{-1}$.

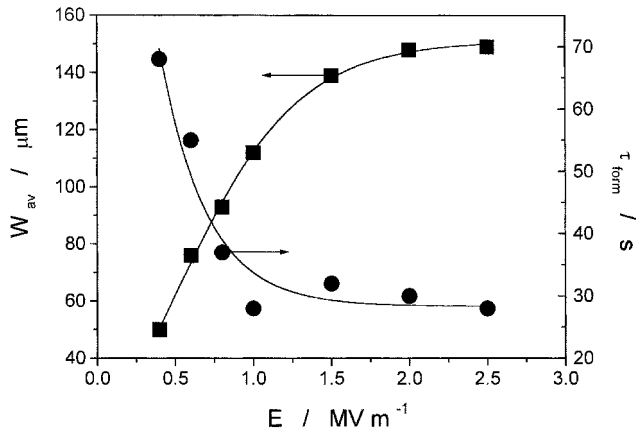


Figure 7. Dependence of the average domain width W_{av} (squares) and time of domain formation τ_{0rm} (circles) on the applied electric field amplitude. Experimental conditions: $10 \mu\text{m}$ cell, 200 Hz square wave field, $T_{AC} - T = 0.5 \text{ K}$.

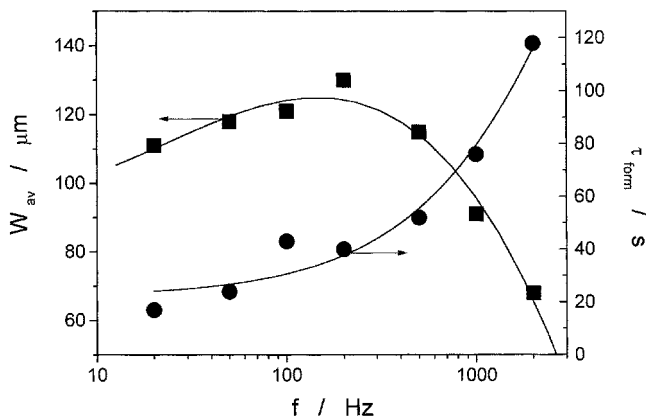


Figure 8. Frequency dependence of the average domain width W_{av} (squares) and time of domain formation τ_{0rm} (circles). Experimental conditions: $10 \mu\text{m}$ cell, $E = 1 \text{ MV m}^{-1}$, square wave field, $T_{AC} - T = 0.5 \text{ K}$.

temperatures close to the transition from SmC^* to the high temperature phase. Below a certain threshold field, in our case approximately 0.2 MV m^{-1} , no smectic layer rearrangement or domain formation can be induced.

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