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Dynamics of the smectic layer reorientation of ferroelectric liquid crystals

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Detailed experimental results of a systematic investigation of the dynamics of the in-plane smectic layer reorientation in SmC* ferroelectric liquid crystals on application of different types of asymmetric electric fields are reported. The reversible reorientation of smectic layers is characterized as a function of field asymmetry, electric field amplitude, frequency, cell gap and temperature. On the basis of the observed behaviour we discuss a phenomenological interpretation of the smectic layer reorientation in terms of dominant influences—director switching, convection due to ionic motion and liquid crystal substrate interactions—which limit the rotation to the amount of twice the tilt angle.

1. Introduction

For the in-plane reorientation of smectic layers, as it will be discussed in this work, a so-called horizontal chevron domain structure is induced by the application of symmetric electric fields of sufficient amplitude. A general introduction and a more detailed account of this process is found in ref. [1]. Andersson *et al.* [2, 3] and Nakayama *et al.* [4] have demonstrated a continuous smectic layer rotation in the smectic A* phase of a ferroelectric liquid crystal material. Rotations of up to several hundred degrees were observed, leading to electric field induced rotational instabilities [3]. They reported the layer reorientation direction to depend on the asymmetry of the field and be continuous above a certain threshold. For ferroelectric [5] and antiferroelectric [6, 7] liquid crystals, the smectic layer rotation has been reported to be reversible, exhibiting thresholdless and threshold behaviour, respectively, with no restriction in the rotation angle observed. It was found that the FLC layer reorientation could not be induced by asymmetric rectangular fields [5], in contrast to our observations. In the present study we have investigated the dynamics of the smectic layer reorientation of a ferroelectric liquid crystal in monostable cells (with one preferred direction of alignment) which is reversible but not continuous (limited by the amount of the tilt angle θ), with respect to electric field asymmetry, amplitude and frequency as well as cell gap and temperature for different time and amplitude asymmetric waveforms.

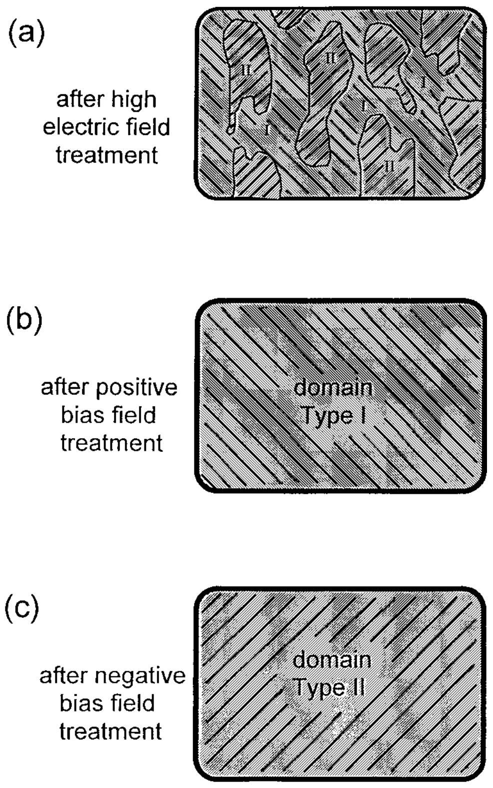

Figure 1. Schematic illustration of the smectic layer reorientation. (*a*) After application of a suitable *symmetric* electric field, the horizontal chevron domain structure is formed; (*b*) after further application of a positively biased or respective time asymmetric field, domains of type II reorient into the structure of domains of type I; (*c*) reversal of the bias polarity or asymmetry of the electric field causes a rearrangement of the smectic layers into domain type II. The sign of this reorientation process is dependent on the configuration (chirality) of the mesogenic molecule.

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2. Experimental

General experimental conditions such as liquid crystal material, cells and equipment used were the same as reported in ref. [1]. As the layer rotation time τ_{layer} we define the time needed for one domain type of a virgin horizontal chevron sample to reorient into the second domain type. In this way, influences through domain nucleation processes can be avoided. Virgin horizontal chevron samples were obtained by subjecting cells that had been cooled from the cholesteric phase to 1 K into the SmC* phase to a 200 Hz symmetric square wave field of amplitude $E = 4\,\mathrm{MV\,m^{-1}}$. This procedure was carried out before each individual measurement.

For a variation of the field asymmetry or bias a 200 Hz electric field of amplitude $E = 4\,\mathrm{MV\,m^{-1}}$ was applied to 6 μm samples at $T_{AC} - T = 1$ K, with T_{AC} being the transition temperature from TGBA* to SmC*. Electric field dependent measurements were carried out on 6 μm samples at $T_{AC} - T = 1$ K with frequency $f = 200$ Hz and 20:80 time asymmetry or $1\,\mathrm{MV\,m^{-1}}$ bias, as well as varying electric field amplitude at constant bias field. The frequency dependence was investigated by using an $E = 4\,\mathrm{MV\,m^{-1}}$ electric field amplitude, 20:80 time asymmetry or $1\,\mathrm{MV\,m^{-1}}$ bias at $T_{AC} - T = 1$ K. The dependence of smectic layer switching on cell gap and temperature was studied for analogous external parameters; frequency $f = 200$ Hz, amplitude $E = 4\,\mathrm{MV\,m^{-1}}$,

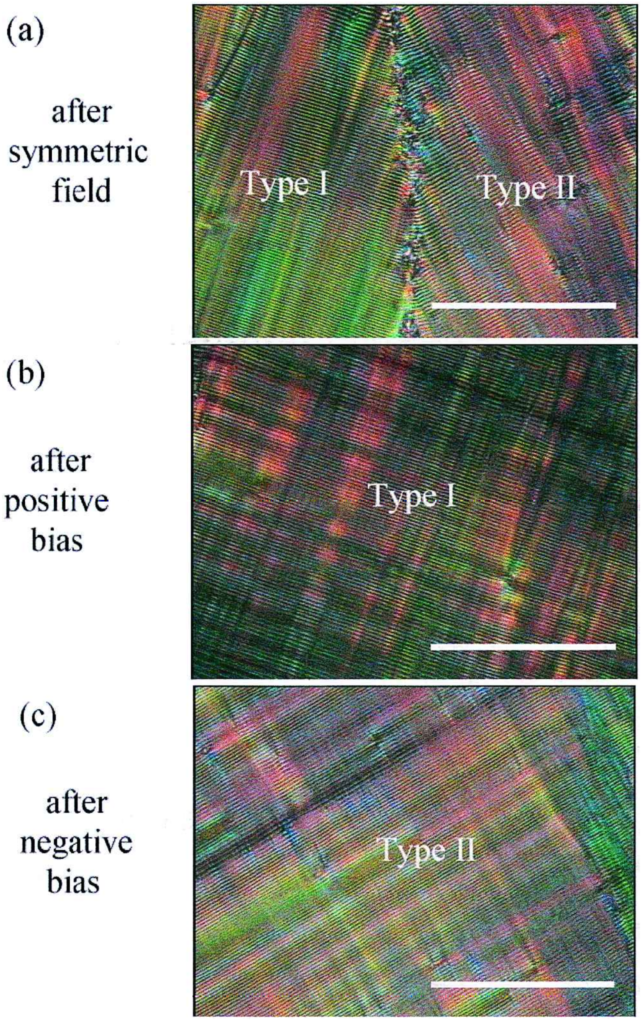

Figure 2. Texture photographs illustrating the schematic behaviour discussed in figure 1; helix lines indicate the direction of the smectic layers. (*a*) Horizontal chevron domains after application of a *symmetric* square wave field; (*b*) rotated smectic layer structure after additional application of a positive bias component; (*c*) rotated layer structure after reversal of the bias field polarity. The bar is equal to 100 μm.

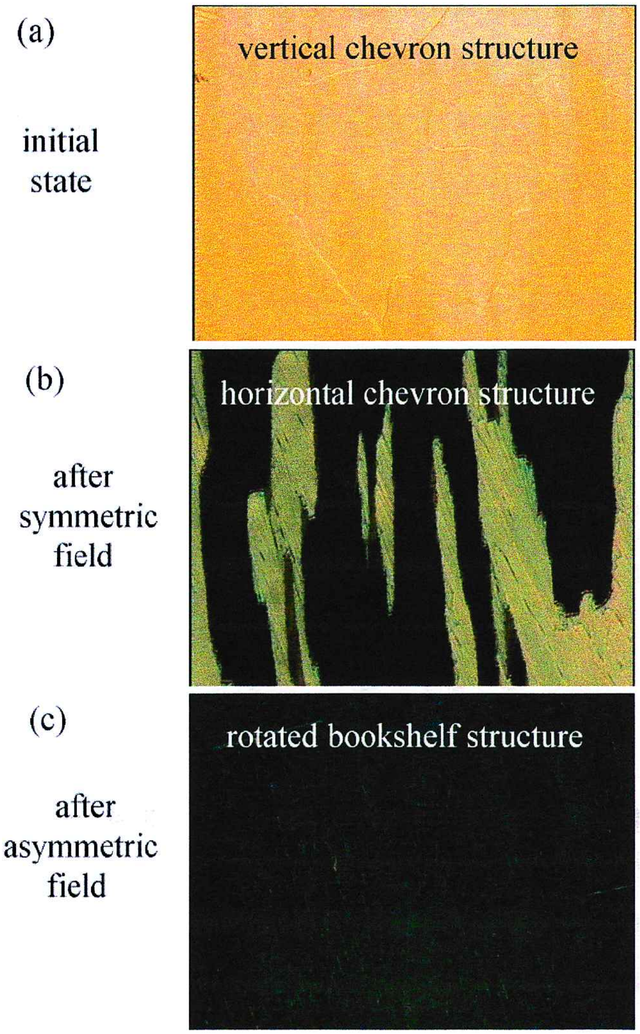

Figure 11. Texture photographs illustrating the behaviour outlined schematically in figure 10. (*a*) Initial director configuration; (*b*) field induced horizontal chevron structure; (*c*) rotated bookshelf structure after application of a biased electric field.

time asymmetry 20 : 80, bias 1 MV m^{-1} at temperature $T_{AC} - T = 1$ K or cell gap $d = 6\,\mu m$, respectively.

3. Experimental results and discussion

As for earlier reports, we also observed that the direction of the smectic layer reorientation is dependent on the time asymmetry of the electric field and the direction of the bias field. In contrast to other reports though [5], we find the layer rotation of ferroelectric liquid crystals to be restricted to values of twice the director tilt angle. The angle α between the smectic layer normal and the rubbing direction is equal to the director tilt angle [8]. During the layer reorientation, the smectic layer normal switches from an angle $+\alpha$ to $-\alpha$ (or $-\alpha$ to $+\alpha$), and thus we observe a reorientation by 2θ. This smectic layer reorientation is reversible by changing the time asymmetry or the bias direction of the electric field. Figure 1 schematically depicts the layer reorientation by application of different bias fields. The texture photographs of figure 2, illustrate the behaviour outlined during

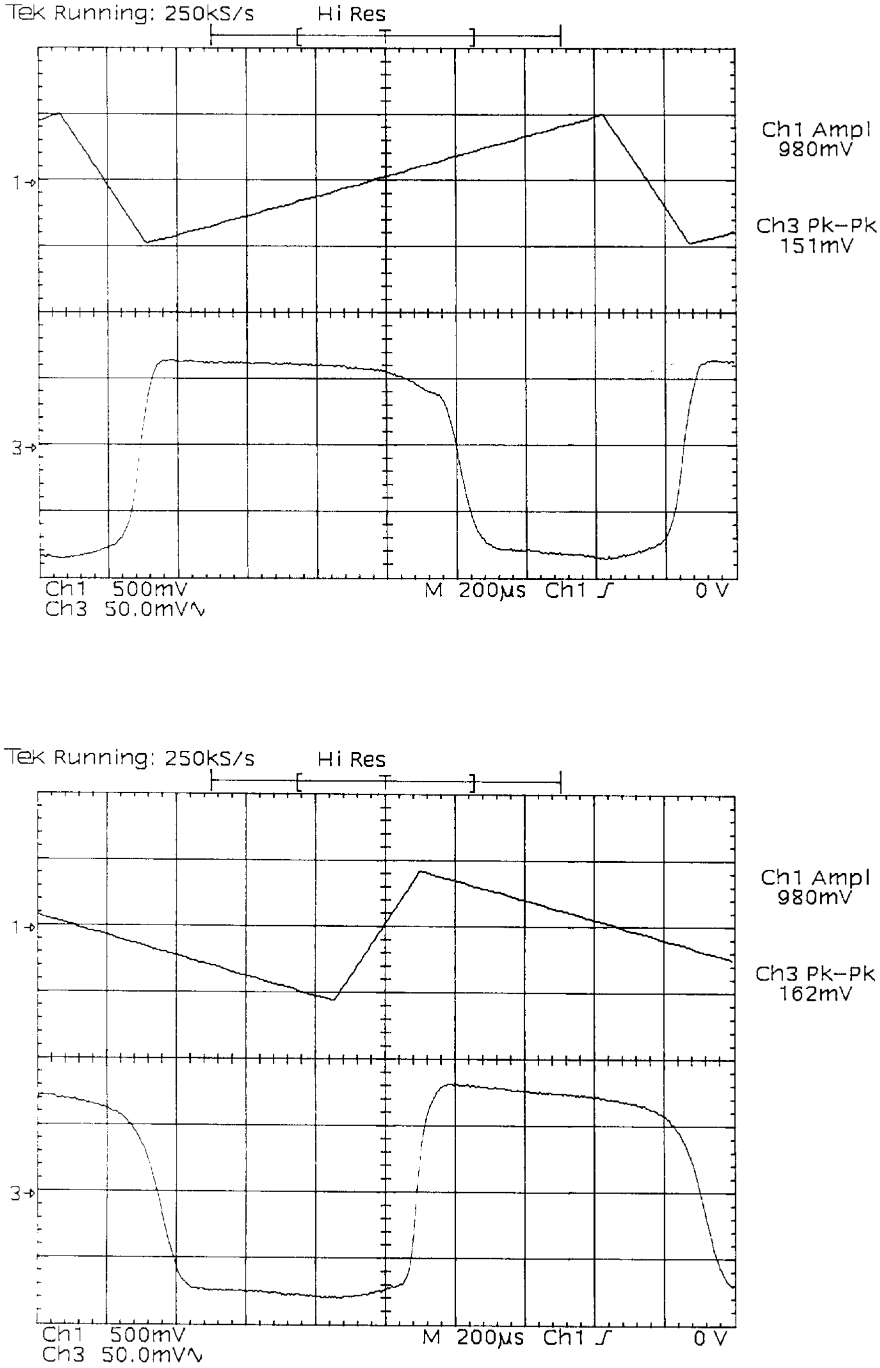

Figure 3. Electro-optic behaviour corresponding to the two states of rotated smectic layers between crossed polarizers with the rubbing direction oriented at an angle of 22·5° with respect to one of the polarizer directions. The electro-optic response is observed to revert, when the direction of asymmetry of the applied electric field (or polarity of the bias) is reversed.

the smectic layer reorientation. After application of a *symmetric* electric a.c. field, a domain structure of horizontal chevrons is formed with smectic layers tilted by the amount of the director tilt angle θ to either side of the rubbing direction [figures 1 (*a*) and 2 (*a*)]. Additional application of a negative bias voltage results in the layer rotation of domain type I to the smectic layer configuration of domain type II [figures 1 (*b*) and 2 (*b*)]. Reversal of the bias field direction causes a layer reorientation of domain type II to domain type I [figures 1 (*c*) and 2 (*c*)]. The line pattern of the texture photographs, due to the helical structure of the SmC* phase, illustrates the direction of the smectic layers. The correponding reversal of the electro-optic behaviour of the two respective domain types is depicted in figure 3 for samples switched between crossed polarizers with the rubbing direction oriented at 22·5° from one of the polarizer directions. The optical response after application of an asymmetric sawtooth voltage is reversed by reversing the applied asymmetry.

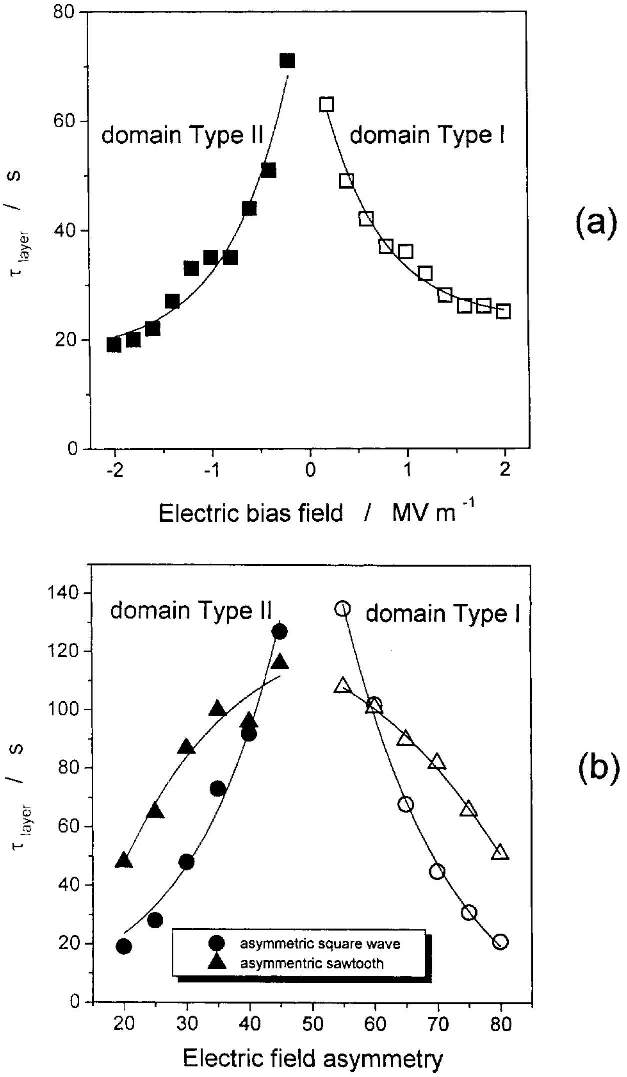

Figure 4. Smectic layer reorientation time τ_{layer} as a function of (*a*) electric bias field and (*b*) time asymmetry (20 means time asymmetry ratio 20 : 80; analogously 80 means time asymmetry ratio 80 : 20, corresponding to a reversal of time asymmetry). Experimental conditions: 6 μm cell, frequency $f = 200$ Hz, amplitude $E = 4$ MV m^{-1}, reduced temperature $T_{\text{AC}} - T = 1$ K.

The switching time needed for the smectic layers to transform from one to the other domain type is strongly dependent on the bias and the asymmetry of the applied electric field, as depicted in figure 4 (*a*) and 4 (*b*), respectively. The layer reorientation times diverge on approaching non-bias or symmetric (asymmetry 50 : 50) field conditions. Reversal of the bias polarity or field asymmetry (for example 20 pos. : 80 neg. corresponding to 80 pos. : 20 neg.) causes opposite domains to reorient their smectic layer structure. In this case the smectic layer response time is symmetric about $E_{\text{bias}} = 0$ V m^{-1} and field asymmetry 50 : 50 (symmetric fields). During further experimental investigations it has become clear that the layer reorientation is a rather complicated process, depending not only on external conditions as characterized in this work. The phase polymorphism [9], ionic contamination [10] and alignment conditions [11] also play an important role. From a first theoretical model it seems that the electroclinic effect cannot be the sole cause of the layer reorientation, but that friction forces too are of great importance. The theoretical studies of the smectic layer reorientation will be reported elsewhere.

A net molecular reorientation per switching cycle can be observed by polarizing microscopy where, depending on bias polarity or asymmetry, one domain type grows at the expense of the less favoured one. This reorientation

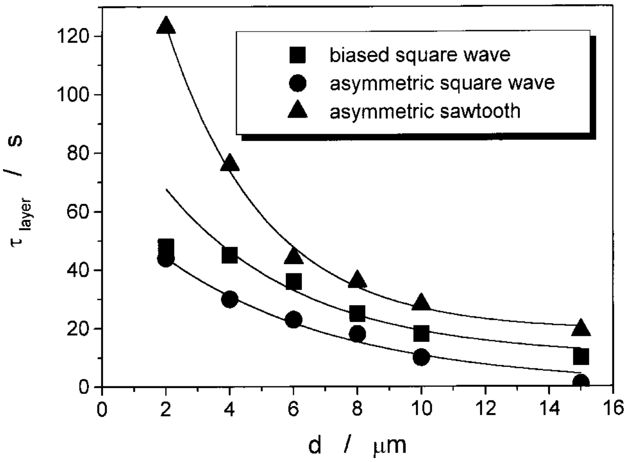

Figure 5. Cell gap dependence of the smectic layer reorientation time for the three different electric waveforms under investigation. Experimental conditions: frequency $f = 200$ Hz, amplitude $E = 4$ MV m^{-1}, asymmetry ratio 20 : 80, bias 1 MV m^{-1}, reduced temperature $T_{\text{AC}} - T = 1$ K.

is directly seen when defect lines between domains of opposite layer tilt are approaching each other and vanish on contact. For samples with strong convective flow, only a rather turbulent mass transport is observed during the reorientation. Strong monostable boundary conditions imposed by rubbed alignment layers of the substrates play an important role in restricting the layer reorientation to twice the amount of the director tilt angle. Figure 5 depicts the layer reorientation time τ_{layer} as a function of cell gap *d*; it is found to decrease with increasing gap for all three waveforms under investigation. This behaviour can readily be understood in terms of the vanishing influence of the substrate alignment layers for thick samples, leaving more liquid crystal material free to reorient in the bulk. It can be seen that the asymmetric square wave is the most efficient waveform for inducing smectic layer rotation, whereas reorientation times for the asymmetric sawtooth are clearly longer. The electric field dependence of the smectic layer reorientation time is depicted in figure 6 (*a*) for the time asymmetric waveform and in figure 6 (*b*) for the biased square wave. τ_{layer} is found to decrease with increasing field amplitude E_{AC} in all cases. Considering the director switching as an important factor for the layer reorientation, it is clear that larger a.c. and bias voltages result in larger asymmetry per switching cycle, especially close to the transition into the TGBA* or N* phase. Also for the field amplitude variation, the time asymmetric square wave is the more efficient for reorienting the smectic layers as compared with the asymmetric sawtooth waveform. It should be mentioned that a smectic layer reorientation can be observed even if the bias field strength is chosen such that only a unipolar pulse voltage is applied to the sample.

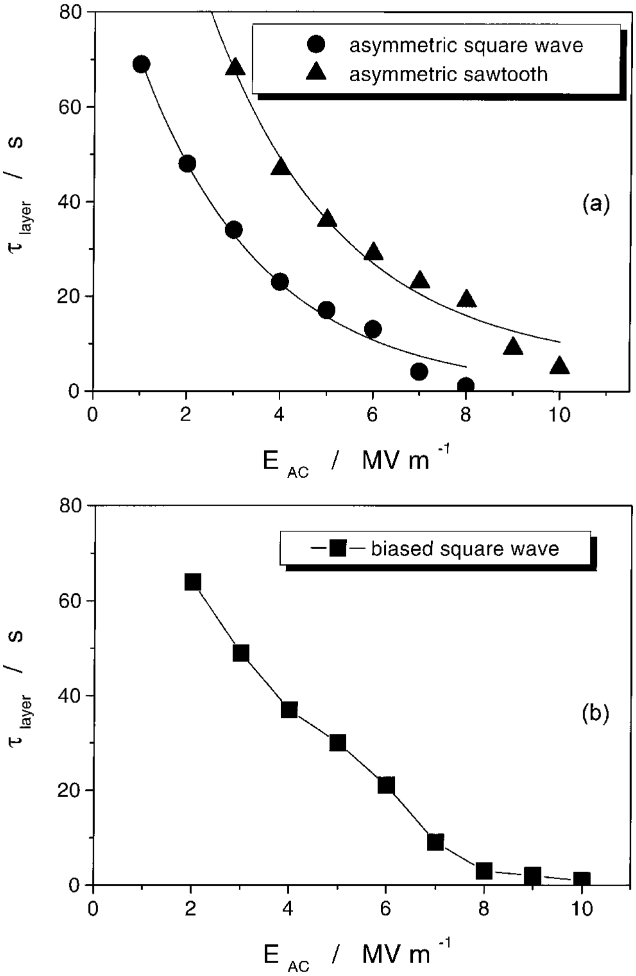

Figure 6. Dependence of the layer reorientation time on applied electric field strength for the three qualitative waveforms studied (*a*) time asymmetric fields and (*b*) at constant bias. Experimental conditions: 6 μm cell, frequency $f = 200$ Hz, time asymmetry 20:80, bias 1 MV m^{-1}, reduced temperature $T_{\text{AC}} - T = 1$ K.

The frequency dependence of the smectic layer reorientation response is somewhat more complicated, as depicted in figure 7. For high frequencies *f*, in the order of several tens of kHz, rather long reorientation times are observed, decreasing with decreasing frequency until at approximately 2 kHz an increase is observed for still further decreasing frequency. At about 100 Hz the layer reorientation times are again found to decrease strongly with decreasing frequency. This dependence is due to several relaxation processes within the liquid crystalline material. The short layer reorientation times observed for low frequencies, smaller than approximately $f = 100$ Hz, are caused by convective flow due to ionic motion. As some mass transport is needed for the smectic layer rearrangement, this strong molecular mobility naturally favours short reorientation times. Approaching

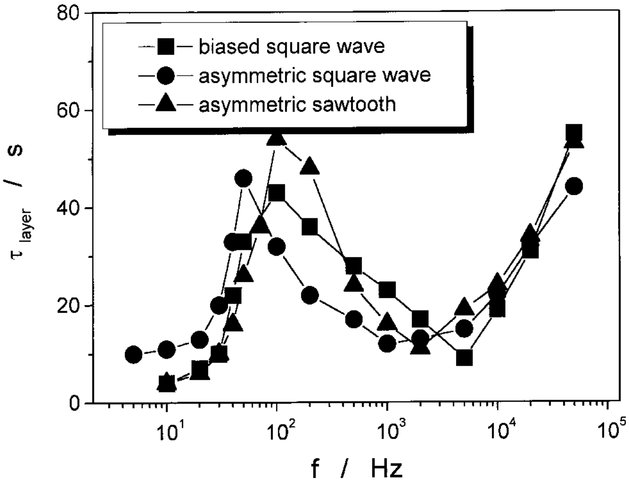

Figure 7. Frequency dependence of the reorientation time of smectic layers for the three waveforms studied. Experimental conditions: 6 μm cell, amplitude $E = 4$ MV m^{-1}, time asymmetry 20:80, bias 1 MV m^{-1}, reduced temperature $T_{\text{AC}} - T = 1$ K.

$f = 100\,\mathrm{Hz}$, the ions are no longer able to follow the electric field and a maximum in $\tau_{\mathrm{layer}}(f)$ is observed. The subsequent decrease of reorientation times is due to the larger number of switching cycles per second at constant electric field. Approaching frequencies in the order of 1 kHz, saturated director switching is observed to cease (by electro-optic methods), decreasing the molecular motion essential for the smectic layer reorientation, and $\tau_{\mathrm{layer}}(f)$ is observed to increase. For frequencies in the range several tenths of kHz, all switching relaxes, observable in diverging layer reorientation times. This interpretation of the frequency dependence of the smectic layer rotation can be demonstrated in the low frequency region by application of a bias field of half the amplitude of the a.c. switching field, resulting in a unipolar pulse voltage and thus trapping ions at the electrodes. Figure 8 depicts the corresponding behaviour. In contrast to the observed decrease in $\tau_{\mathrm{layer}}(f)$ for bias fields of $E_{\mathrm{bias}} = 1\,\mathrm{MV\,m^{-1}}$, we observe the expected increase, due to the lower number of switching cycles per second, if ionic motion is prohibited.

Figure 9 depicts the temperature dependence of the layer reorientation time which is observed to increase with decreasing temperature. This behaviour may be attributed to the increasing viscosity when lowering the temperature into the smectic C* phase. Also here we do observe shorter reorientation times for time asymmetric square wave fields as compared with corresponding sawtooth fields.

The field induced reorientation of smectic layers can also be used to align smectic C* samples in a uniform bookshelf geometry. Considering samples without helical superstructure, vertical chevrons are transformed into horizontal chevrons with rotated uniform bookshelf layers with respect to the rubbing direction, by subjecting the cell to a symmetric voltage. Then, by application of an asymmetric field, the vertical chevron defects of the horizonal chevron domain structure are removed through a reorientation of the smectic layers. The procedure is schematically depicted in figure 10 (*a*)–(*c*) and corresponding texture photographs are shown in figure 11. The quality of the alignment after the electric field treatment, as compared with the virgin texture before any electric field application, is demonstrated in figure 12 for the normalized transmitted light intensity between crossed polarizers as a function of the sample rotation angle ϕ, showing a very good improvement of the dark state at $\phi = 0$, and thus improved contrast and viewing characteristics.

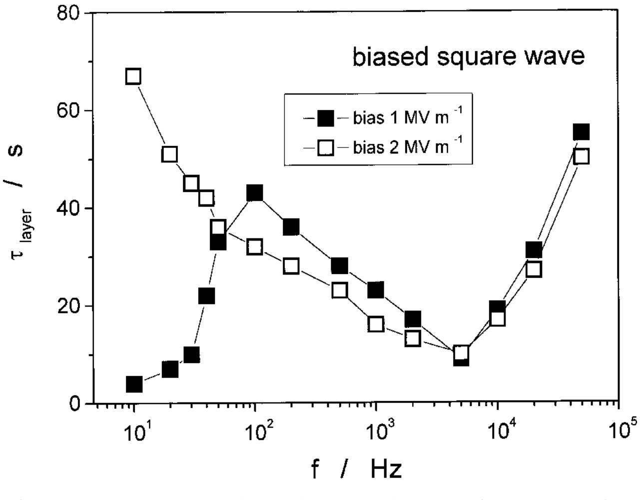

Figure 8. Frequency dependence of the smectic layer reorientation time for different applied bias fields. Trapping ions at the electrodes (bias $2\,\mathrm{MV\,m^{-1}}$ at applied field of $4\,\mathrm{MV\,m^{-1}}$, thus applying a unipolar pulse voltage) results in increasing reorientation times as compared with simply biased switching ($1\,\mathrm{MV\,m^{-1}}$) for frequencies smaller than $f = 100\,\mathrm{Hz}$ (see text).

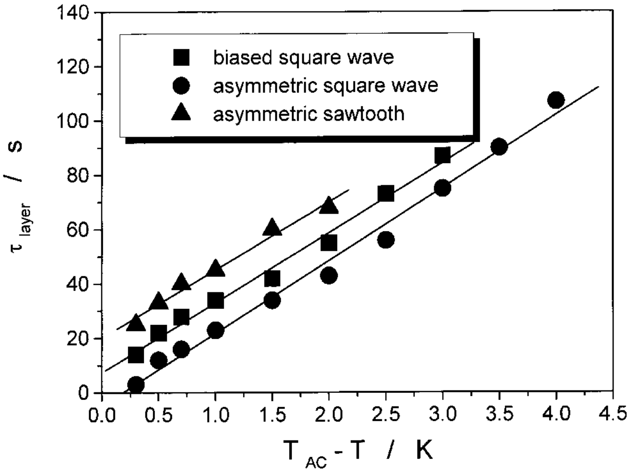

Figure 9. Dependence of the smectic layer reorientation times on reduced temperature $T_{\mathrm{AC}} - T$ below the TGBA*–SmC* phase transition for the three waveforms investigated. Experimental conditions: 6 μm cell, frequency $f = 200\,\mathrm{Hz}$, amplitude $E = 4\,\mathrm{MV\,m^{-1}}$, time asymmetry 20:80, bias $1\,\mathrm{MV\,m^{-1}}$.

4. Conclusions

The dynamics of the smectic layer reorientation under asymmetric electric fields was systematically investigated with respect to asymmetry ratio, bias field, electric field waveform and amplitude, frequency, cell gap and temperature. The times needed for one horizontal chevron domain type to reorient its smectic layer orientation into that of the second domain type, τ_{layer}, are found to:

(a) increase with decreasing asymmetry and bias voltage,
(b) decrease with increasing electric field amplitude,

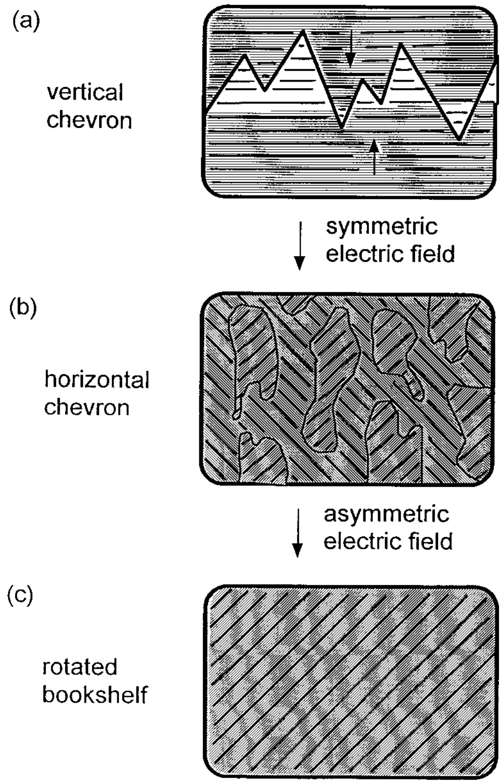

Figure 10. Schematic illustration for obtaining a well oriented smectic C* rotated bookshelf texture. (*a*) Initial condition is a vertical chevron structure with zigzag defects. Smectic layers are oriented perpendicular to the rubbing direction and make an angle with the substrate normal; (*b*) after application of a symmetric electric square wave field of suitable amplitude, a horizontal chevron domain structure is formed with smectic layers of the bookshelf type tilted with respect to the rubbing direction by the amount of the director tilt angle; (*c*) application of an asymmetric or biased electric field transforms the horizontal chevron domain structure into a rotated bookshelf structure with the smectic layer normal inclined to the rubbing direction by the amount of the director tilt angle—if this procedure is carried out with non-helical SmC* materials, a well oriented bookshelf texture is obtained.

(c) increase with decreasing temperature below the transition point into the SmC* phase,
(d) decrease with increasing cell gap.

The frequency dependence of the layer reorientation is somewhat more complicated and discussed in detail. From the experimental data three major factors dominating the layer reorientation can be extracted:

(1) Strong liquid crystal–substrate interactions, due to rubbed polyimide alignment layers limit the reorientation to twice the director tilt angle; it is expected that for weak LC–substrate interactions the rotation becomes continuous. A systematic study of this question is being carried out at the present time.

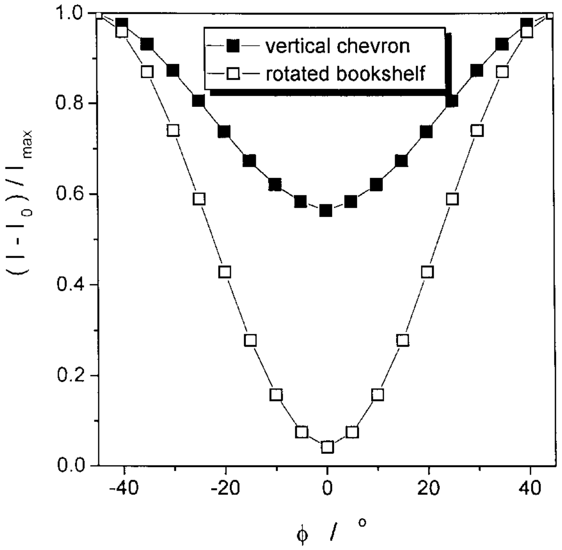

Figure 12. Measurement of the normalized transmitted light intensity as a function of the rotation angle ϕ of the cell between crossed polarizers for the vertical chevron structure before electric field treatment (closed symbols) and after the procedure outlined schematically in figure 10 in the rotated bookshelf geometry (open symbols). The dark state at rotation angle $\phi = 0$, corresponding to the direction of rubbing, is clearly enhanced, leading to greater contrast.

(2) The director switching process, consisting of both ferroelectric and electroclinic contributions, is of great importance for the reorientation of smectic layers.

(3) For the smectic layer rearrangement, molecular motion provided by the switching of the director is essential. Reorientation times are greatly enhanced by additional material flow due to convection caused by large ionic mobility at low frequencies and large electric field amplitudes.

Further studies on the concentration effect of ionic impurities on the smectic layer reorientation will be reported in due course.

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