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Domain growth in a ferroelectric liquid crystal with horizontal chevron layer geometry

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Abstract. The motion of domain walls of horizontal chevron interfaces was investigated with respect to externally applied conditions. As in solid state ferroelectrics, a linear displacement of domain walls is observed with time, thus leading to a constant domain wall velocity. Variation of the electric field time asymmetry ratio, which is the driving force of the domain wall motion, leads to a linear increase in domain wall velocity, just as does an increase of the electric field amplitude. The frequency dependence of the domain wall velocity is strongly influenced by motion of ionic impurities and ceasing of saturated director switching, while the temperature dependence is mainly governed by viscosity effects of an Arrhenius-like behaviour.

1. Introduction

Domain growth under application of external fields is known for a variety of different systems, the most well known probably being ferroelectric materials [1, 2], such as BaTiO₃, or ferromagnetic materials [3], such as iron. At small or moderate fields, the reorientation of the spontaneous polarization P_S or magnetization M , respectively, does not occur by rotation of whole domains but rather through a displacement of domain walls separating regions of different orientation of P_S or M . The process is generally related to nucleation and growth of the energetically favoured domains at the expense of the unfavoured ones.

Similarly, domains of opposite direction of the spontaneous polarization can be observed in chiral smectic liquid crystals, when the intrinsic helical superstructure is suppressed by boundary conditions [4], the so called bookshelf geometry, with smectic layers perpendicular to the substrate plates. Especially in the smectic C* phase, which in addition to the orientational order of the long molecular axis also exhibits a one-dimensional positional order of the molecules' centres of mass (layers), the direction of the spontaneous polarization can easily be reversed by application of electric fields. Within the smectic layer plane the molecules are isotropically distributed, thus representing a two-dimensional liquid. Generally, the smectic layer geometry observed is more complicated than the ideal bookshelf arrangement and exhibits the *vertical* chevron geometry, with layers being tilted with respect to the substrates [5]. Regions of opposite layer inclination are mediated by defects called zigzag lines [6].

Another smectic layer arrangement is the so-called *horizontal* chevron geometry, first demonstrated by Patel and Goodby [7] by cooling across a N*–SmC* transition under DC field conditions. Alternatively, these structures can also irreversibly be induced from a vertical chevron or an unoriented configuration by application of a symmetric electric field of sufficient

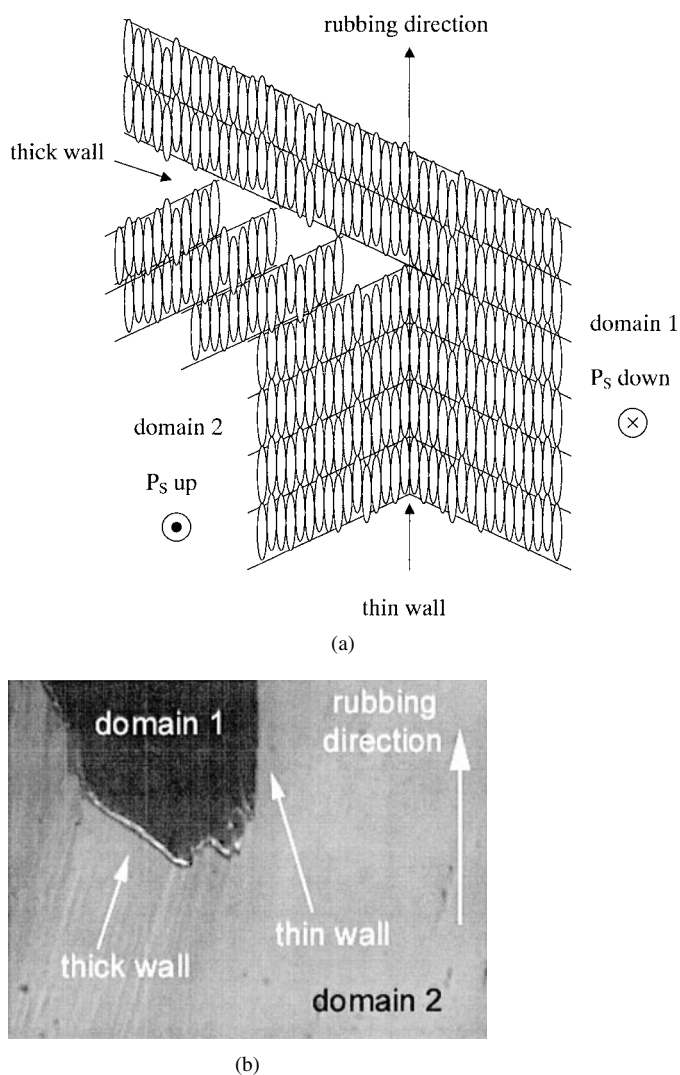


Figure 1. (a) Schematic illustration of the horizontal chevron domain structure with thick and thin domain walls mediating regions of opposite layer inclination. (b) Respective texture, transmission micrograph between crossed polarizers. Image size is $290\ \mu\text{m}$ times $220\ \mu\text{m}$.

amplitude in the SmC^* phase [8], as done in this study. The smectic layers in different domains are perpendicular to the substrate planes (bookshelf), but with the layer normal inclined to either side of the direction of monostable boundary condition (rubbing direction), as schematically illustrated in figure 1(a). In the field-free state the director in both domains is oriented along the rubbing directions, thus the two domain types exhibit a spontaneous polarization in opposite directions, pointing into or out of the substrate plane. Regions mediating domains of opposite layer inclination are domain walls, which can be thin, without layer discontinuities, or thick, exhibiting discontinuities of smectic layers. Figure 1(b) shows a microscopic texture photograph of the domain structure (under DC bias to distinguish both domain types) between crossed polarizers.

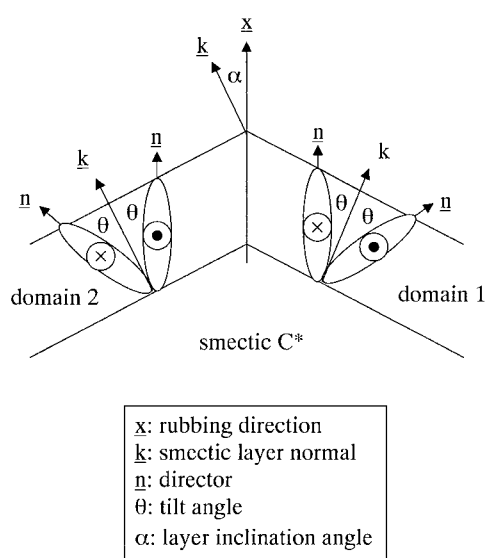


Figure 2. Schematic illustration of polarization reversal and director switching in horizontal chevrons.

At this point we should stress that we will not follow the domain wall motion (motion of zigzag lines) during polarization reversal by application of a *symmetric* AC electric field. Experimental investigations [9, 10] of this kind as well as computer simulations [11–13] have been reported over recent years. In ferroelectric smectic liquid crystals, being anisotropic fluids, this process is very fast, of the order of several microseconds. In this study we will investigate the growth of horizontal chevron domains as illustrated in figure 1, by application of *asymmetric* AC electric fields. This process is rather slow, of the order of several seconds, while the superimposed reversal of the spontaneous polarization in this case is only a secondary effect and is schematically depicted in figure 2. Molecules in each domain are confined to the tilt cone of opening angle θ with respect to the smectic layer normal. Reversal of the field direction causes a reversal of the direction of the spontaneous polarization, combined with a motion of the director along the tilt cone into the opposite stable orientation. This reorientation process, generally studied on application of a *symmetric* electric field, does *not* cause a displacement of horizontal chevron domain walls. In contrast, we will here study the growth of one domain type with smectic layers inclined by an angle α , often equal to the director tilt angle θ , with respect to the rubbing direction. The energetically favoured domain type (direction of domain wall motion) can be selected by the choice of the electric field asymmetry, depending on the sign of the spontaneous polarization and the electroclinic coefficient.

2. Experiment

The ferroelectric liquid crystal used in this study is a commercially available epoxy compound, 4-[(S,S)-2,3-epoxy-hexyloxy]phenyl-4-decyloxybenzoate, from Aldrich. Its polymorphism on cooling was given by: Iso. 95 N* 76.9 TGBA* 76.5 SmC* 54 SmI* 39 Cryst. (temperatures in °C). The SmI* phase is monotropic and the occurrence of the narrow frustrated twist grain boundary phase is of no direct relevance to the topic under investigation, as we will confine our interest to the ferroelectric SmC* phase only. Cells were prepared by spin-coating ITO

covered glass substrates with a polyimide, subsequent rubbing and cell assembly at a cell gap $6 \mu\text{m}$ and parallel, monostable boundary conditions. The liquid crystal was then introduced by capillary action in the isotropic phase.

The horizontal chevron domain structures were obtained by cooling into the SmC^* phase ($T_C - T = 1.5 \text{ K}$) and application of a *symmetric* electric square wave field of amplitude $E = 4 \text{ MV m}^{-1}$ at frequency $f = 200 \text{ Hz}$ by a Philips PM5138 function generator in combination with a F20A power amplifier from FLC Electronics. The domain wall motion was induced by application of time *asymmetric* square wave fields and followed by video recording of the transmission microscopic image between crossed polarizers (Leitz). Image analysis was performed by use of standard software (Adobe Premiere). The error bars on the measurement curves refer to uncertainties in the determination of the domain wall position. They cannot refer to pinning effects of domain walls due to impurities within the liquid crystal or substrate roughness. Nevertheless, pinning has been avoided, as the domain wall motion could be followed *in situ* by polarizing microscopy and pinning can readily be seen. During the measurements the temperature of the cell was controlled within $\pm 0.1 \text{ K}$ by a Mettler FP80 temperature controller and a FP82 hot stage.

General experimental conditions for domain wall motion were as follows: liquid crystal cell gap $6 \mu\text{m}$, electric field amplitude $E = 4 \text{ MV m}^{-1}$, electric field time asymmetry 20% positive/80% neg. square waveform (selecting growth of domain 1), frequency $f = 200 \text{ Hz}$, reduced temperature $T_C - T = 1.5 \text{ K}$. For the different measurement series presented below, only the one respective parameter was varied, leaving others unchanged.

3. Experimental results and discussion

Figure 3 shows a series of texture micrographs taken 1 s apart, during the domain growth process initiated by application of an asymmetric electric field. If starting from a sample solely in domain 2 orientation, growth of domain 1 occurs via nucleation, where the nucleation sites are not predetermined. Increase of the electric field amplitude increases the number of nucleation sites. In all cases it is found that the domain nucleation time is much smaller than the time of domain growth and can thus be neglected. By image analysis and setting the position of the thick domain wall at $t_0 = 0$ to $d_0 = 0 \mu\text{m}$, we can determine the domain wall position as a function of time (figure 4). A linear displacement of the domain wall is observed, from which a constant domain wall velocity of $v_{\text{growth}} = 13.6 \mu\text{m s}^{-1}$ can be determined for the specific field conditions applied. This value corresponds to a domain wall mobility of approximately $\mu = 3.4 \times 10^{-8} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, being small as compared to solid state ferroelectrics ($\mu = 2.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for BaTiO_3 ([1], chapter IV.8), $\mu = 2-8 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for GASH ([1], chapter VIII.2), $\mu = 1.4 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for Rochelle salt ([1], chapter VII.6)), which can be accounted for by different experimental conditions, material parameters and bearing in mind that the domain growth process studied here is somewhat different from the polarization reversal in solid ferroelectric materials. (For a comparison: $P_S \approx 2 \times 10^{-4} \text{ C m}^{-2}$ for the ferroelectric liquid crystal (FLC) and $P_S \approx 0.2 \text{ C m}^{-2}$ for BaTiO_3 , applied fields $E \approx 4 \text{ MV m}^{-1}$ for FLC and $E \approx 0.04 \text{ MV m}^{-1}$ for solid state ferroelectrics. Thus the spontaneous polarization of the liquid crystal is a factor of 10^3 smaller and the applied fields a factor of 10^2 higher in comparison to typical values for solid state ferroelectric materials). Nevertheless, the qualitative behaviour found for the growth of horizontal chevrons in FLCs is similar to that observed for solid state ferroelectrics during polarization reversal [1].

The mechanism of reorientation of smectic layers under asymmetric electric fields is at the present time not fully understood. It is believed that it is caused by the dynamics of the

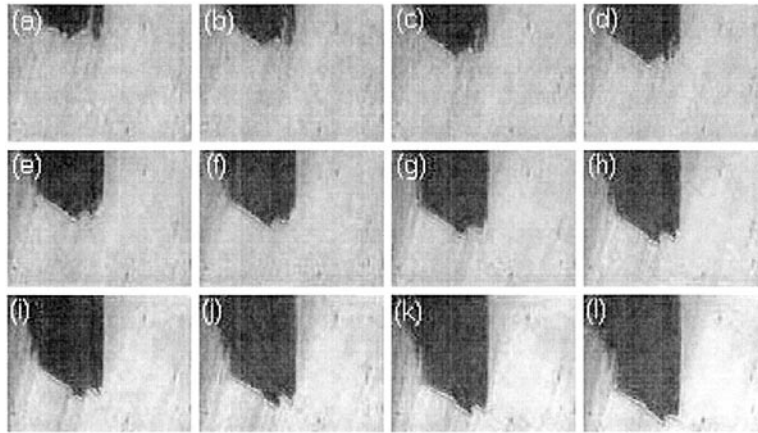


Figure 3. Series of texture micrographs during domain growth, taken 1 s apart. Each image size is $290 \mu\text{m}$ times $220 \mu\text{m}$.

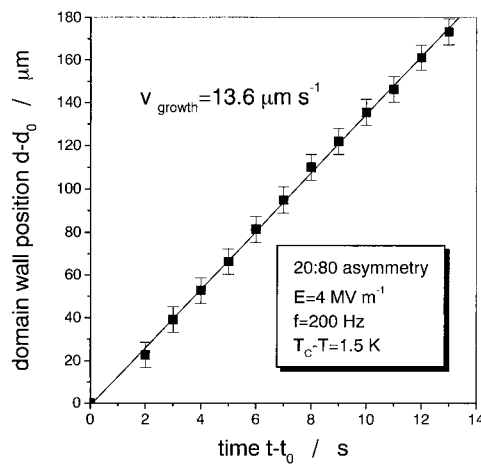


Figure 4. Domain wall position as a function of time of applied asymmetric electric field. From the linear relationship a domain wall velocity of $v_{\text{growth}} = 13.6 \mu\text{m s}^{-1}$ is obtained for the specific experimental conditions applied.

director switching, including electroclinic contributions and effects of ionic motion, maybe bearing some similarities to a Lehmann-like rotational phenomenon [14, 15], with the time derivative of the electric field playing the role of the temperature gradient in the original Lehmann effect. It can only be observed in chiral materials and is essentially dependent on an asymmetry of the applied electric field. Figure 5 depicts the dependence of the domain wall velocity on the electric field asymmetry ratio. For scaling on an interval of $[-1, 1]$, the asymmetry ratio is arbitrarily defined as $+(1 - \% \text{negative} / \% \text{positive})$ for fields selecting growth of domain 2 (anticlockwise, right handed rotation) and $-(1 - \% \text{positive} / \% \text{negative})$ for fields selecting growth of domain 1 (clockwise, left handed rotation). An asymmetry ratio of zero thus corresponds to a symmetric electric field. A basically linear dependence of the domain wall velocity on asymmetry ratio is observed. Reversal of the field asymmetry results in a reversal of the direction of domain wall motion.

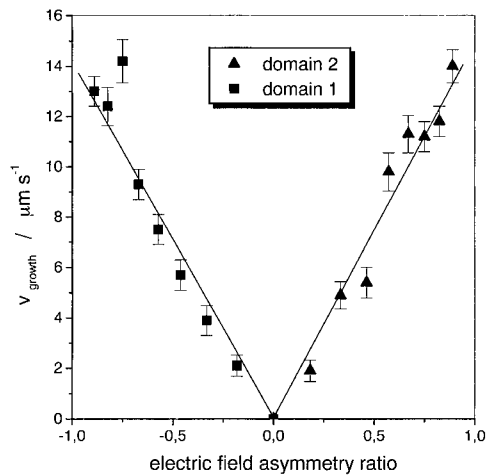


Figure 5. Domain wall velocity as a function of electric field asymmetry ratio. Increasing the time asymmetry of the applied field, a linear increase of v_{growth} is observed. Reversal of the time asymmetry results in a reversal of the direction of domain wall motion.

The electric field amplitude dependence of the domain wall velocity is depicted in figure 6. Following a simple model of domain wall motion in solid state ferroelectrics [16] based on a treatment analogous to a rigid domain wall moving in a viscous liquid, v_{growth} should be linearly dependent on E :

$$v_{growth} = \mu E \quad (1)$$

or

$$v_{growth} = \mu(E - E_0) \quad (2)$$

if a critical field E_0 is observed. Relation (1) is approximately fulfilled in the present study (solid line), while a slightly non-linear fit of the data (dotted line) for moderate field amplitudes of $E < 6 \text{ MV m}^{-1}$ suggests a small critical field of the order of $E_0 = 0.1 \text{ MV m}^{-1}$, which is comparable to the threshold field for director switching. For amplitudes $E > 8 \text{ MV m}^{-1}$ turbulent flow starts to set in. According to (1) the ratio between the domain growth velocity and the electric field amplitude should be constant for a fixed temperature and equal to the wall mobility. From figure 6 a value of $\mu \approx 4 \times 10^{-8} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is determined, comparable to that obtained from figure 4.

Figure 7(a) shows the dependence of the domain wall velocity on the frequency of the applied electric field. Following a basically linear increase, a clear maximum is obtained at frequencies around $f = 1500 \text{ Hz}$. A more comparable presentation is depicted in figure 7(b), with $v_{growth} f^{-1}(f)$ showing the domain wall displacement per switching cycle as a function of frequency. Three regimes can be distinguished: (I) for frequencies $f \leq 150\text{--}300 \text{ Hz}$ the domain wall motion is strongly enhanced by the ionic conductivity of the sample, which is in general accordance with dielectric measurements, showing a relaxation of ionic motion in low molecular mass liquid crystals at approximately 150 Hz. (II) In the range of $200\text{--}300 \text{ Hz} \leq f \leq 1500 \text{ Hz}$ a plateau is reached, indicating a constant domain wall displacement of approximately 30 nm per switching cycle. The upper limit of this range corresponds to the frequency up to which saturated director switching is observed by electro-optic measurements. (III) For frequencies $f \geq 1500 \text{ Hz}$ saturated director switching gradually

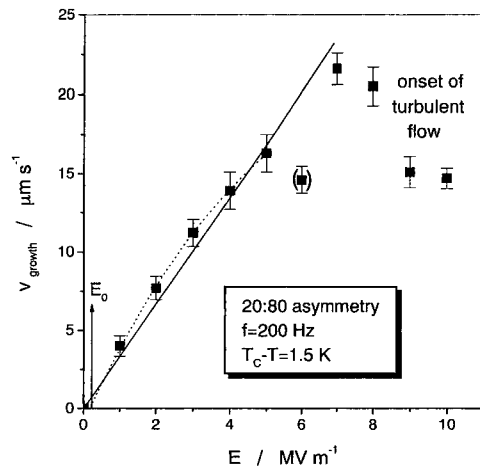


Figure 6. Domain wall velocity as a function of applied electric field amplitude, showing a basically linear increase (solid line). A slightly non-linear fit of the data for moderate amplitudes (dotted line) suggests the existence of a small critical field E_0 of the order of the threshold field for director switching. From the ratio of domain wall velocity to field amplitude a basically constant domain wall mobility of $\mu = 4 \times 10^{-8} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ can be determined.

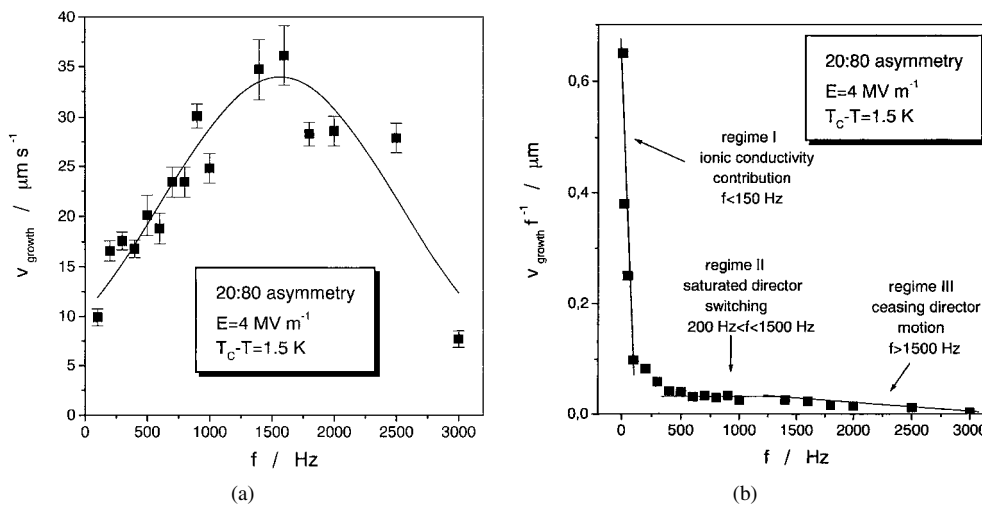


Figure 7. (a) Frequency dependence of the domain wall velocity. (b) From the ratio $v_{growth} f^{-1}$ (domain wall displacement per switching cycle), as a function of frequency, three distinct regimes can be observed: (I) ionically dominated growth for low frequencies ($f < 150 \text{ Hz}$), (II) constant domain wall displacement per switching cycle for medium frequencies ($200 \text{ Hz} < f < 1500 \text{ Hz}$) and (III) vanishing domain growth as the director reorientation ceases for high frequencies ($f > 1500 \text{ Hz}$).

ceases and for even larger frequencies ($f > 10 \text{ kHz}$) also the electroclinic effect relaxes and domain wall motion vanishes completely.

Approaching the transition to the high temperature phase, an exponential increase of the domain wall velocity is observed. As v_{growth} is proportional to the domain wall mobility, $v_{growth} \sim \mu$, and this in turn is inversely proportional to the viscosity, $\mu \sim 1/\eta$, the observed

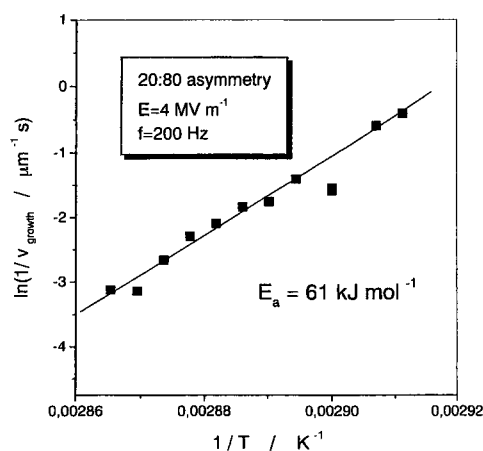


Figure 8. The temperature dependence of the domain wall velocity exhibits a strongly increasing v_{growth} as the transition to the high temperature phase is approached at T_C . An Arrhenius-like representation reveals that the temperature dependence of the domain wall velocity is mainly caused by the viscous behaviour of the liquid crystal and yields an activation energy of $E_a = 61 \text{ kJ mol}^{-1}$.

behaviour can be attributed to the temperature dependence of the viscosity, which for liquid crystals often exhibits an Arrhenius-like behaviour [17]:

$$\eta = \eta_0 \exp(E_a/k_B T). \quad (3)$$

A respective plot of $\ln(1/v_{growth})$ as a function of $1/T$ is shown in figure 8 and yields an activation energy for the domain wall displacement of $E_a = 61 \text{ kJ mol}^{-1}$, which is in good accordance with values determined for liquid crystals from viscosity measurements or dielectric spectroscopy.

4. Conclusions

The motion of domain walls in horizontal chevron structures of a ferroelectric SmC* liquid crystal has been investigated with respect to electric field asymmetry, amplitude, frequency and temperature. Domain walls were found to move uniformly with a velocity v_{growth} , which is linearly increasing with increasing electric field asymmetry and amplitude until turbulent flow sets in. Three distinct regimes are observed in the frequency dependence of the domain wall velocity: at low frequencies the domain wall motion is strongly influenced by ionic contributions, while at medium frequencies, where saturated director switching is observed, the displacement per switching cycle is constant, then at high frequencies the domain wall motion gradually vanishes with ceasing director reorientation. The temperature dependence of the domain wall motion is basically determined by the viscosity, showing an Arrhenius-like behaviour.

Acknowledgments

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