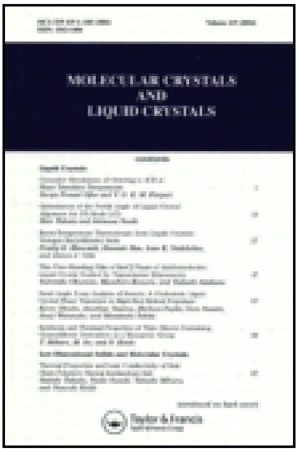

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SPONTANEOUS PERIODIC IN-LAYER DIRECTOR MODULATION IN TILTED CHIRAL SMECTICS

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Abstract Periodic director modulation within the smectic layers is observed as 1D-stripe or 2D textures in free suspended films in SmC*, SmC^*_A and SmI^*_A phases in a temperature range where the helix along the smectic layer normal is spontaneously unwound. Temperature changes of the texture are significantly different in all three studied phases. Applied electric field affects the periodicity and the orientation of the stripe texture. Models of both 1D and 2D textures are presented.

INTRODUCTION

Recently the existence of periodic director modulations propagating within the smectic layers has been reported[1,2] in the ferroelectric (FE) and antiferroelectric (AF) SmC* phase. The in-plane modulated structure, which is a direct consequence of the symmetry of chiral tilted smectics, is formed in free suspended films of materials in which the helix along the smectic layer normal (z-helix) is spontaneously unwound. When approaching the temperature where the z-helix is unwound, the modulated structure appears abruptly behind a phase front as a stripe texture.[1,2] The stripes, visible in polarized and non-polarized light, are formed by arrays of defects. Two systems of defects have been found, near the upper and lower surfaces of free suspended film, which are mutually shifted by half of the stripe periodicity.

The observed defects have been interpreted as a system of π-walls terminated by π-disclinations at a definite distance $h<D/2$ (D is the film thickness) from the film surface. Between the walls the director modulations take place.[1]

The in-plane modulations are supposed to be governed by the elastic chiral term[3] $D_1(rot c)_z$, where z is parallel to the smectic layer normal. The azimuthal angle φ defines the orientation of director $\boldsymbol{n} = (\sin\theta \cos\varphi, \sin\theta \sin\varphi, \cos\theta)$, θ is the tilt angle. The projection of $\boldsymbol{n}$ on the smectic layer is a vector $\boldsymbol{c} = (\cos\varphi, \sin\varphi)$. An analysis of the elastic term showed[4] that the in-plane modulation should be accompanied by a regular array of defects (disclinations or walls), at which the angle φ changes discontinuously, which is in accordance with the observed texture.

A phenomenological description of the observed structure taking into account the elasticity of the SmC* phase has been worked out,[1] which gives a linear dependence of the stripe periodicity, d, on the film thickness. This dependence was confirmed by the experiment.

In this contribution, properties of the in-plane modulated structure are studied in the FE and AF SmC* phases and in the AF SmI* phase. An attention is paid to the evolution of the structure in temperature changes and its behavior in electric fields.

EXPERIMENTAL

The liquid crystals used in the study were *Cln/m*, which exhibit the helix twist inversion in the FE SmC* phase,[5] *MHPOBC* with the helix twist inversion in the AF SmC* phase[6] (SmC^*_A) and *BI8/6* with the helix twist inversion in the AF SmI* phase[7,8] (SmI^*_A). In all these materials the helix twist inversion phenomenon is accompanied by the z-helix unwinding in a broad temperature range containing the inversion temperature T_i. The sequences of phases and the temperature ranges where the in-plane modulations exist are shown in Figure 1 for all studied compounds.

In experiments free suspended films 5-150μm thick were used. The film thickness was determined either as the distance between focusing lower and upper surfaces in microscopic measurements, or from the value of rotatory power. The textures were studied by polarizing microscope and the periodicity of the modulation determined from the diffraction of laser light (630nm). The electric field was applied in the suspended film plane.

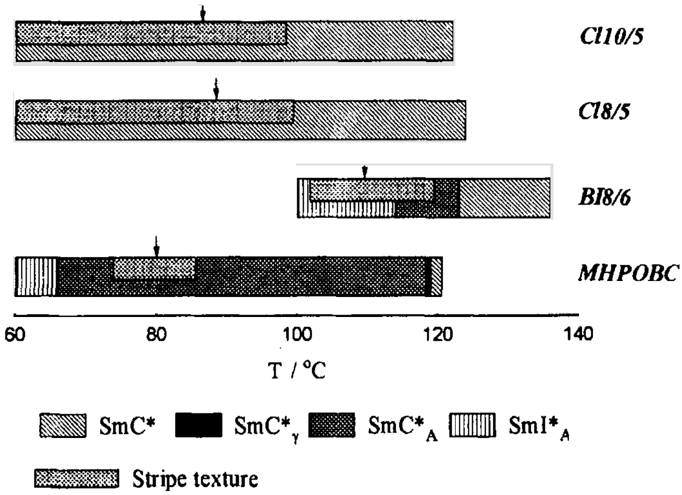

FIGURE 1 The sequences of phases and the temperature ranges of the existence of the stripe texture for studied compounds. The arrows indicate the inversion temperatures T_i.

RESULTS

Far above the inversion temperature T_i a typical homeotropic texture is observed. On cooling, in the vicinity of T_i the stripe texture abruptly appears in all studied materials and persists when the temperature is lowered (see Figures 2a and 3). As it has been pointed out in Ref. 2., no stripe texture occurs in films of the thickness below 10μm.

The periodicity, *d*, of the stripe texture depends on the temperature, the dependence being different for the SmC*, SmC*$_A$ and SmI*$_A$ phases. In Figure 4 critical temperature behavior of the periodicity wave vector in the FE phase $q=2\pi/d$ is shown. It is seen that this behaviour is more expressed for thinner samples. For *BI8/6*, *q* exhibits an anomaly around the SmC*$_A$ - SmI*$_A$ phase transition (Figure 5), for the SmC*$_A$ phase of *MHPOBC* *q* is temperature independent (see inset in Figure 5).

Under an applied d.c. electric field the stripe periodicity steeply decreases with the increased field for both FE and AF phases (see Figures 6a and b). Above a critical field, which is ~60V/mm for FE and ~500V/mm for AF phases, the stripe texture disappears because the in-plane modulation is unwound.

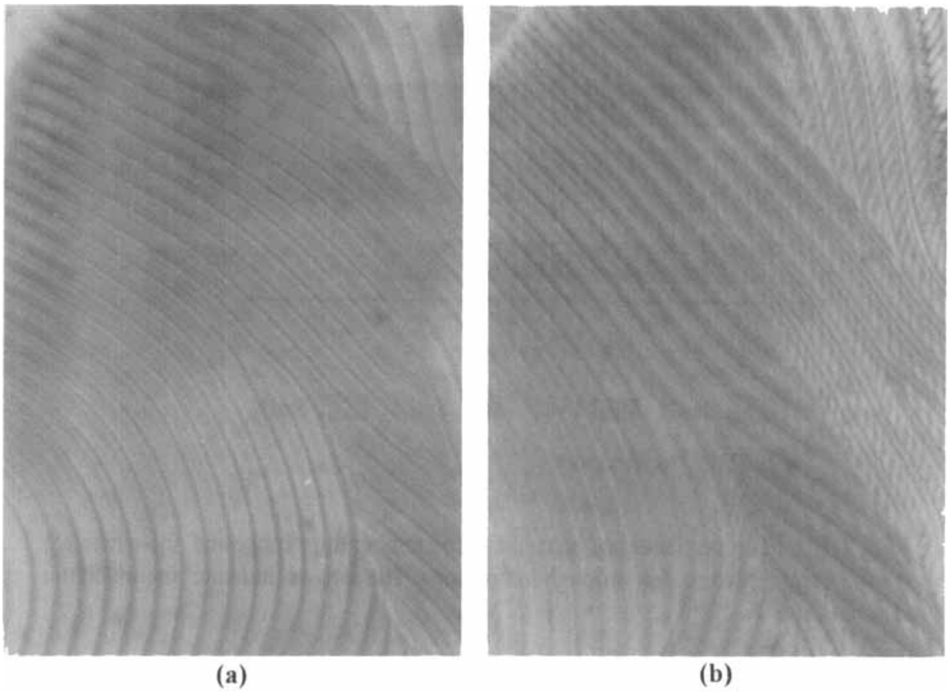

FIGURE 2 The stripe texture in the SmC* phase of *Cl8/5* at temperatures: (a) 98.5°C, only primary lines are seen, (b) 98.0°C, the secondary lines are visible. (See Color Plate II).

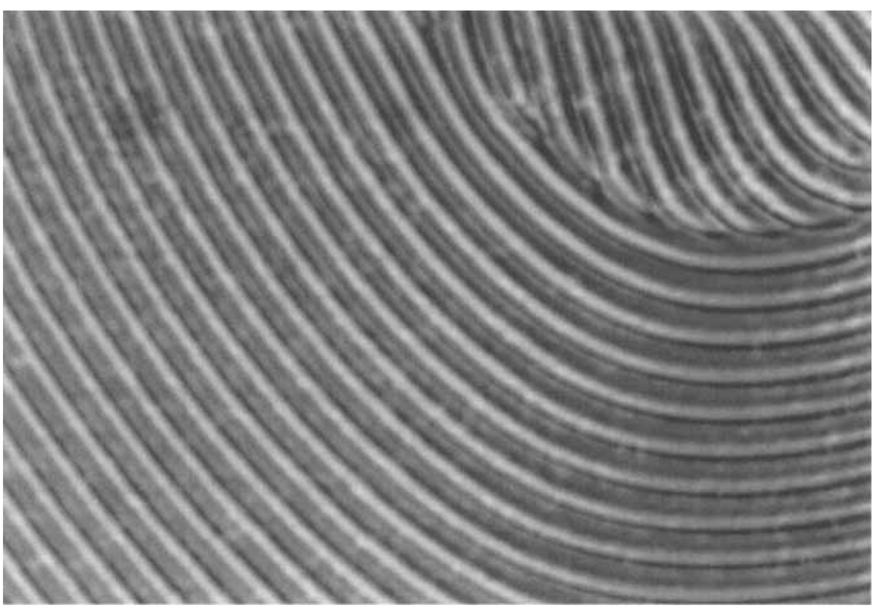

FIGURE 3 The stripe texture in the SmI*$_A$ phase of *B18/6*. (See Color Plate III).

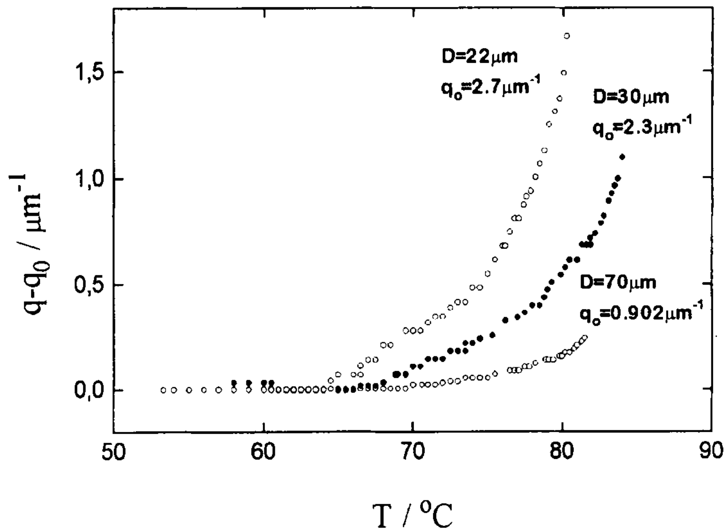

FIGURE 4 The temperature dependence of the critical part of the stripe periodicity wave vector q-q_0 for *Cl8/5* and different film thickness. q_0 is a noncritical part of the wave vector.

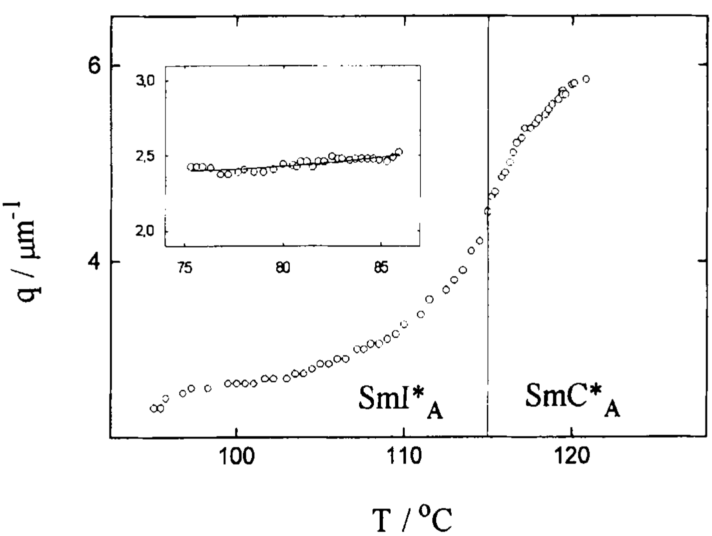

FIGURE 5 The temperature dependence of the stripe periodicity wave vector for *BI8/6*, in the inset for *MHPOBC*.

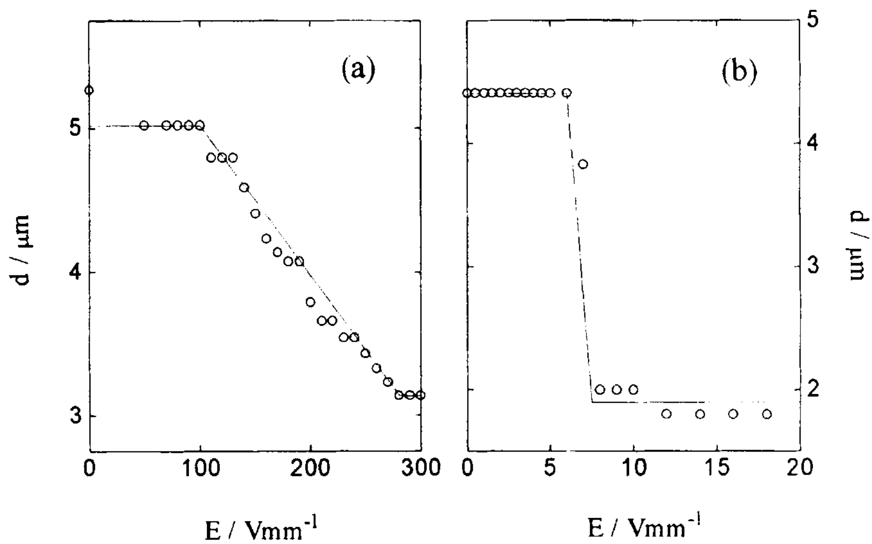

FIGURE 6 The stripe periodicity versus the applied electric d.c. field for (a) *MHPOBC*, (b) *C110/5*.

Besides changing the stripe periodicity, the external electric field has also a reorientation effect on the stripes. At d.c. field or low frequency fields, the stripes become oriented perpendicularly to the field direction. Under the fields of about 100 Hz the stripes become parallel to the field direction (see Figures 7a and b).

Under the d.c. field the stripe texture is induced even at temperatures above the temperature range where the stripes spontaneously arise. In that case, they become oriented perpendicularly to the field direction (Figure 8).

With films thicker than about 20μm, an additional system of lines can appear on cooling, which are inclined by a definite angle towards the primary lines (see Figure. 2b). The inclination of the secondary lines with respect to the primary ones is opposite when focused near upper and lower surfaces, which corresponds to their equal orientation under the film reversion.[1] It should be noted that the inclination of secondary lines is reversed for the opposite enantiomer.

In still thicker samples more complicated textures are observed, namely hexagonal and square textures. Microscopic observations show that, similarly as the stripe patterns,

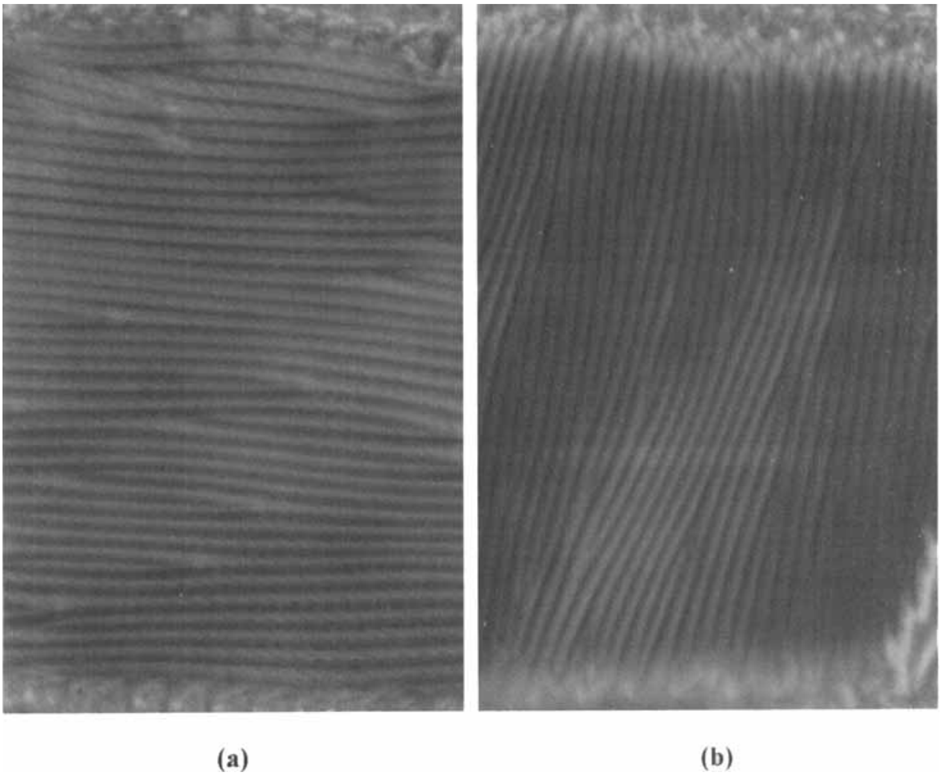

FIGURE 7 The stripe texture in *Cl10/5* at 76°C under the electric field of 10V/mm applied vertically from the sample edges, which are decorated by an array of special defects. (a) d.c. field, (b) a.c. field of the frequency 100Hz. (See Color Plate IV).

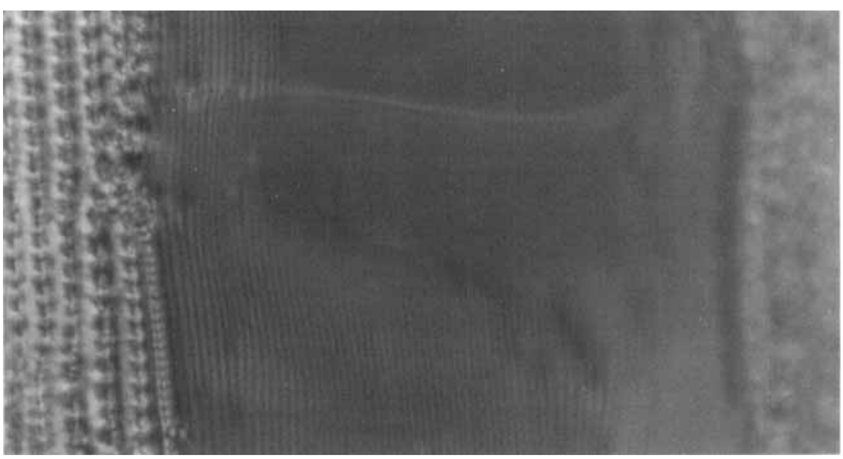

FIGURE 8 The stripe texture in *Cl8/5* induced by d.c. electric field of 5 V/mm applied horizontally. (See Color Plate V).

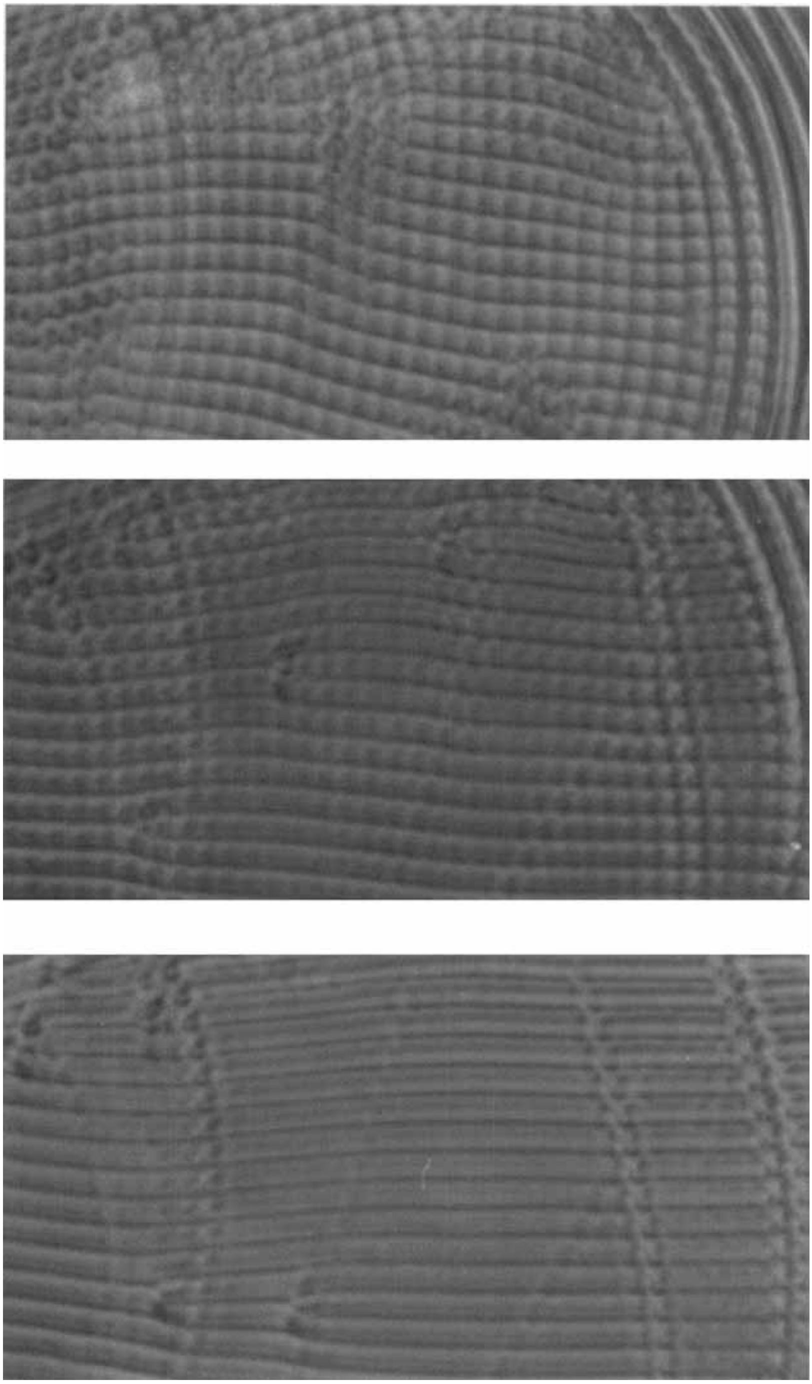

FIGURE 9 The square-stripe texture transformation under the applied electric field in *MHPOBC* free suspended film at 85°C. $E = 0$; 75 and 95 V/mm, top - down. (See Color Plate VI).

the square patterns focused at upper and lower surfaces are mutually shifted. Upper square corners are located above centres of the squares. The contrast on the hexagonal texture is more complicated and does not allow to recognize the upper and lower textures by refocusing. Under an application of a d.c. electric field both square and hexagonal textures are simplified to the stripe texture (see Fig. 9).

DISCUSSION AND CONCLUSIONS

The experimental results show that the existence of spontaneous in-plane director modulations is connected with the spontaneous unwinding of the z-helix. We proved it for the SmC*, SmC^*_A and SmI^*_A phases. The modulations are allowed by the symmetry of these phases, but probably the coexistence with the z-helix is conflicting. On the other hand, when the z-helix is unwound by an external field, the in-plane modulation is usually also unwound. Only near above the temperature range of the spontaneous z-helix unwinding, the electric field that unwinds it is low enough to allow the existence of in-plane modulations. In this way, in-plane modulations can be induced by the external electric field out of the temperature range of its spontaneous existence.

A critical film thickness for the appearance of the in-plane modulations is well defined. In thin films more simple (1D) modulations occur, at higher film thickness, 2D patterns (double stripes, squares, hexagons) are formed. This is valid in all studied phases.

In the SmC* phase the observed critical temperature dependence of the stripe periodicity shows slightly first order transition from uniform to the stripe phase. In the SmC^*_A phase the temperature independence of the stripe periodicity shows a strong first order transition. The anomaly detected around the SmC^*_A - SmI^*_A phase transition may be due to a difference in elastic properties between both phases.

In Fig. 10 a possible director field corresponding to the simple stripe texture is presented. It is supposed that between the walls at one surface the director rotates changing the azimuthal angle φ by π, the rotation being opposite at upper and lower surfaces. The rotation of director field is governed by the chiral term $D_l(rot c)_z = D_l \cos\varphi \, d\varphi/dx$ with x-axis perpendicular to walls and $\varphi=\varphi(x, z)$. A detailed analysis of possible

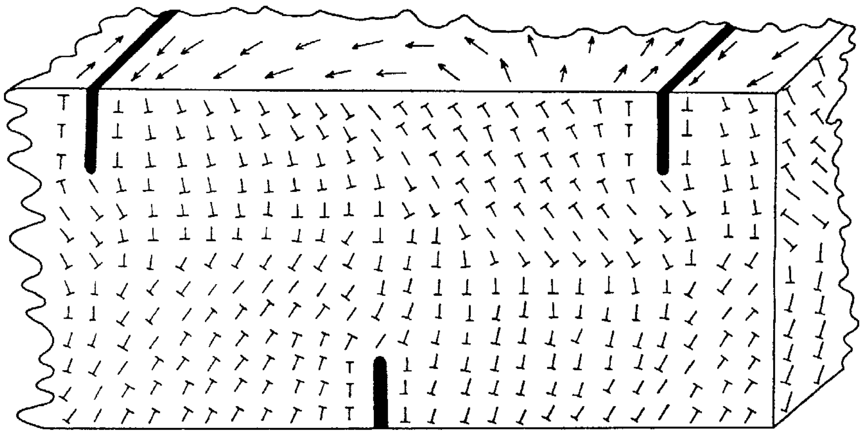

FIGURE 10 The director modulation in stripe texture of free suspended film. Directors are represented by nails, the point of which is directed to the observer. On the upper surface the *c*-directors are shown. Walls near the upper and lower surfaces are represented by thick bars.

modulated surface domains in dependence of chirality is presented in Ref. 1. The domains at the opposite surfaces have to be connected by a director twist along the layer normal. In this model the π-disclinations that terminate walls near the opposite surfaces have the same sign. Their repulsing is responsible for the mutually shifted positions of walls near the surfaces.

A model for the square texture is presented in Figure 11, where a possible director field in one square is shown. Squares are limited by π-walls, the *c*-vector orientation on which follows from the analysis of the term $D_l(rot c)_z = D_l \cos\varphi\, d\varphi/dx + \sin\varphi\, d\varphi/dy$. Near three square corners there are $(-\pi/2)$-disclinations, at one corner there is $(2\pi-\pi/2)$-disclination, all disclination being perpendicular to the film plane. An asymmetrical director field in the squares is in accordance with the observed texture (see the top of Fig. 9). A complete surface structure is obtained by translation of squares in two dimensions. As it is observed in the experiment, the structure is shifted at two opposite surfaces. The upper and lower surface structures can be hardly joined continuously. They have to terminate at a complicated wall in the film bulk. This is probably the reason, why this type of structure occurs only in thick films.

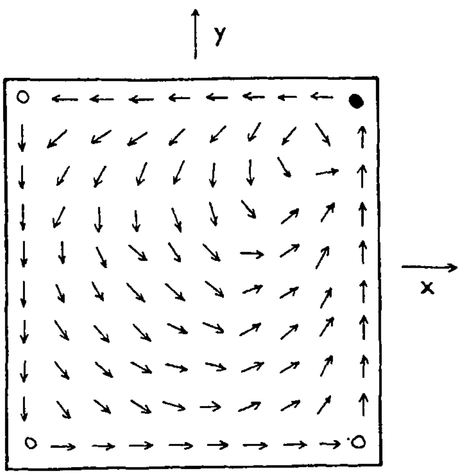

FIGURE 11 The surface structure of the square texture represented by *c*-directors. The full lines are π-walls, the open circles are cross sections of (-π/2)-disclinations, the full circle is a cross section of (2π-π/2)-disclination.

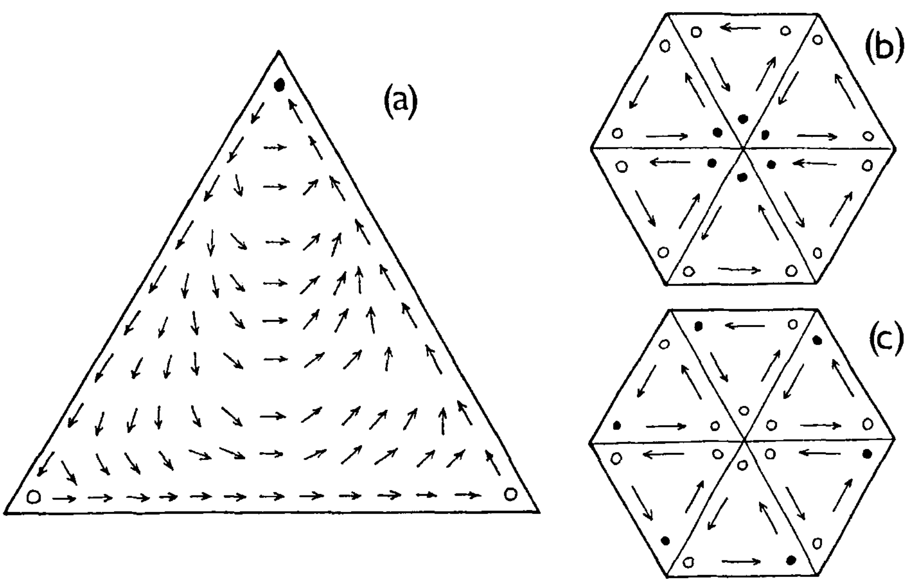

FIGURE 12 The surface structure of the hexagonal texture. (a) *c*-directors in one triangle. The full lines are π-walls, the open circles are cross sections of (-2π/3)-disclinations, the full circle is a cross section of (2π-2π/3)-disclination. (b) and (c) are two ways how to tile triangles to a two dimensional structure.

A model for the hexagonal texture is shown in Figures 12a, b, c. It is composed of triangles limited by π-walls, with (-2π/3)-disclinations in two corners and (2π-2π/3)-disclination in one corner (see Fig. 12a). The triangles can be tiled by two ways to a 2D structure (see Figs. 12b and c).

Models for both square and hexagonal textures are inspired by the model of Hinshaw and Petschek.[4] However, in our model 2π-disclination is shifted from the centre into one corner of the square or triangle, to make the structure consistent with microscopic observations.

The behavior of the in-plane modulations under the applied electric field is driven by the interaction of the local polarization with the field. The texture changes are caused by a lost of structure stability under the field, which can be inferred from the analysis of the elastic free energy. Details will be published elsewhere.

Acknowledgements

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