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Preliminary communication

Modulated structures with field-controlled direction and periodicity in SmC* liquid crystals

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A stable modulated structure in the form of stripes occurs in SmC* liquid crystal cells well below the SmA–SmC* transition point, without any applied or pre-applied electric field. The periodicity of the modulations is much smaller than the cell thickness. The stripes are tilted away from the rub direction of the cell. A low applied voltage changes both the periodicity and orientation of the stripes, allowing for electric field control of optical diffraction at the stripes.

1. Introduction

Ferroelectric chiral smectic C (SmC*) liquid crystals are promising materials for applications in fast electro-optic devices. Extensive studies during the last two decades have advanced the understanding of rather complex structures formed in the so-called surface-stabilized SmC* cells. The idea of surface stabilization is to unwind the helicoidal SmC* structure. It can be achieved when the cell thickness d is smaller than the helicoidal pitch p [1]. Normally, the preparation of an aligned SmC* slab starts with a homogeneously aligned smectic A (SmA) texture at elevated temperatures. The SmA layers are oriented normally to the glass plates that bound the cell. Upon cooling to the SmC* phase, this ‘bookshelf’ structure becomes distorted: the smectic layers tilt in the vertical plane and usually form a wall defect—‘chevron’ [2, 3]. At the tip of the chevron, the layers reverse their tilt. Chevrons are explained as the result of the contraction of layers, compensated by their tilt [2–7]; for more details, see the works by Nakagawa [4], Limat [5] and Vaupotic *et al.* [6]. The layers contraction is caused by molecular tilt (SmC* phase) or by enhanced molecular ordering at low temperatures, so that the chevrons can also form in the SmA phase [8].

Chevron structure is only the first level of defect hierarchy in smectic cells [8–21]. Very often, the chevrons are decorated by focal-conic domains [8]. Sometimes, instead of the usual ‘vertical’ chevrons (the layers tilt in the plane normal to the cell), one observes ‘horizontal’ chevrons [9–11]. The structures become even more

sophisticated when an external field is involved. Several groups have reported on SmC* stripes induced either directly by the field or after the field is removed [12–20]. The periodicity of stripes is of the order of the cell thickness or larger.

We report on a new type of a fine modulated (stripe) structure that forms in comparably thick ($d \geq 10 \mu\text{m}$) SmC* cells. The cells are still thin enough to keep the SmC* helix unwound. The modulations occur without any external field, either imposed or pre-imposed, at temperatures well below the SmA–SmC* transition. The periodicity of modulations is much smaller than d ; the stripes are tilted with respect to the rub direction at the bounding plates; finally, both the periodicity and orientation of the stripes can be controlled by an external electric field.

2. Experimental

We used the SmC* mixture Felix-015/100 (Hoechst, Germany) with the phase sequence: Cr–12°C–SmC*–72°C–SmA–83°C–N*–86°C–I and the following parameters at 25°C (reported by the manufacturer): spontaneous polarization $P_s = 33 \text{ nC cm}^{-2}$, molecular cone angle $2\theta = 51^\circ$ (as measured by applying a strong d.c. electric field to a planar cell), effective cone angle $2\theta_{\text{eff}} = 26.5^\circ$ (as measured optically by using two memory states in a $2 \mu\text{m}$ cell with no electric field applied). The helical pitch in the cholesteric N* phase at 83°C is larger than $100 \mu\text{m}$.

The cells comprised two ITO coated glass plates. The substrates were spin-coated with different alignment materials: polyimide SE-610 (Nissan Chem. Ind.), polyimide DuPont 2555 and polyvinylalcohol (PVA). For a

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typical nematic, pentylcyanobiphenyl (5CB), the director tilt angle (measured from the substrate) is 6° – 9° with SE-610, 1° – 2° with DuPont 2555 and less than 1° with PVA. The substrates were rubbed and cells assembled in either parallel or antiparallel fashion. The liquid crystal was filled by capillary action in the isotropic phase, then slowly cooled.

3. Results and discussion

At high temperatures, the SmA layers are normal to the rub direction. When the SmA phase is cooled, a chevron texture forms with elongated parabolic focal-conic domains that reduce the stress at the chevron tip [9]. The chevron is preserved in the SmC* phase as indicated by zig-zag walls separating domains with opposite orientation of the chevron tip [2].

Cooling to the deep SmC* phase ($\approx 35^\circ\text{C}$, which is about 40°C below the SmA–SmC* transition) results in a modulated stripe structure, figures 1 and 2. The stripes appear if $d \geq 10 \mu\text{m}$; no external electric field is involved. Their period Λ increases with d , remaining noticeably smaller than d , figure 3. The apparent period doubles if the microscope is defocused from the middle plane of the sample. The dependence $\Lambda(d)$ was measured by optical diffraction (see below) for a wedge cell with $10 \mu\text{m} \leq d \leq 27 \mu\text{m}$. The stripes are visible also in very thick samples (up to $75 \mu\text{m}$, which is the limit of good alignment).

Modulations occur in cells with both parallel and antiparallel assembling, regardless of the type of alignment material. Moreover, the modulations occur in both ‘uniform’ and ‘twisted’ chevron states. These two states differ by the type of director dependence on the x coordinate normal to the cell plates; the uniform state

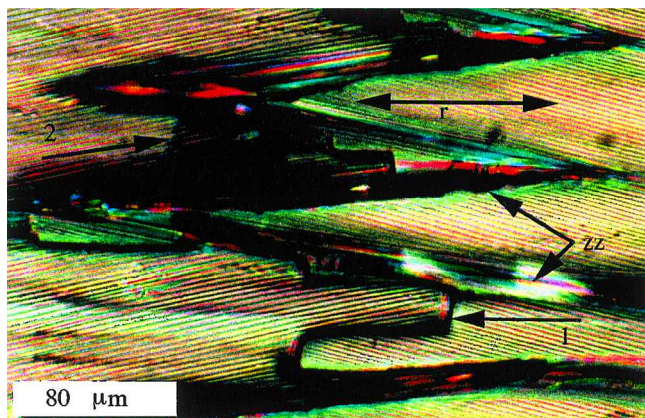


Figure 1. Modulated structure in a SmC* cell of thickness $21 \mu\text{m}$ at 22°C . DuPont 2555 coating, antiparallel rubbing. A highly defectuous zone is chosen to show zig-zag walls (zz), domain walls between stripes in two uniform states (1), and domain walls between twisted and uniform states (2); ‘r’ denotes the rub direction.

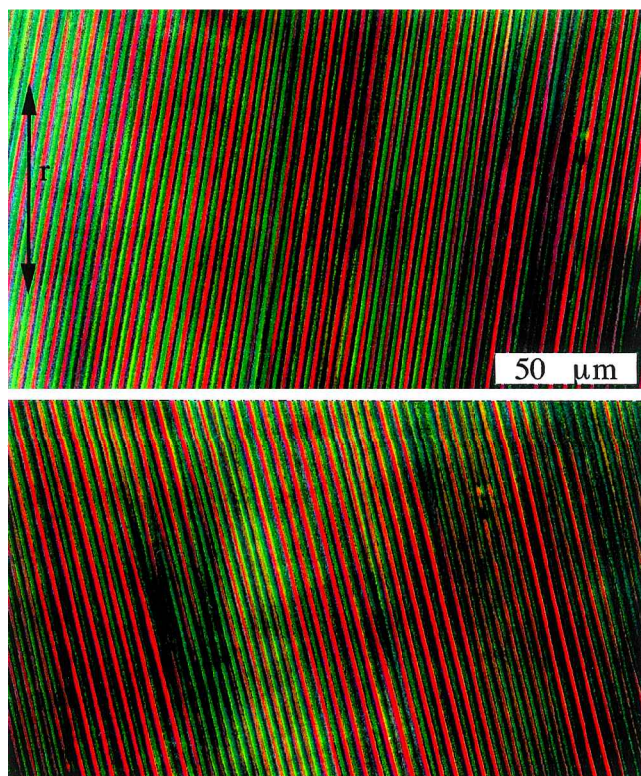


Figure 2. Two orientations of stripes in the SmC* cell ($d = 21 \mu\text{m}$); $U = 0 \text{ V}$ (a) and $U = 0.6 \text{ V}$ (b).

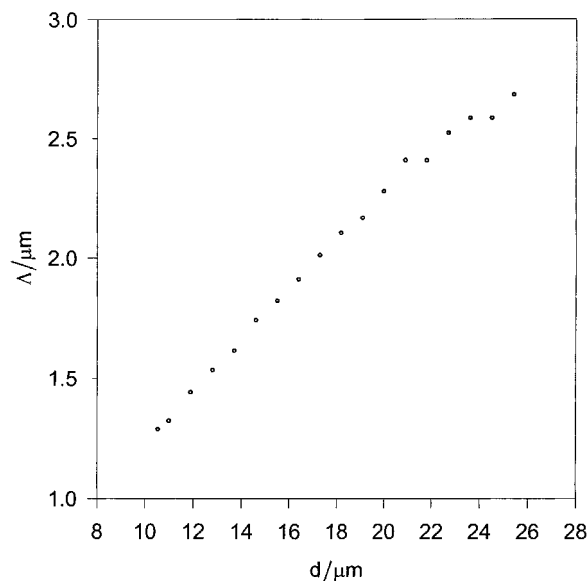


Figure 3. Modulation wavelength Λ as the function of the cell thickness d (PVA coating) at 22°C .

produces extinction textures between crossed polarizers of a microscope, while the twisted state does not. For more details see, e.g. [22]. There are no modulations at the zig-zag walls, where the chevron is destroyed (figure 1).

The ‘uniform’ state produces extinction texture when the molecular projections onto the horizontal plane are

directed along the polarizer or analyser. The stripes are oriented along the extinction directions (i.e. along the average direction of molecular projections) and make an angle $\alpha = \pm(10\text{--}15)^\circ$ with the rub direction. Since the molecules can be tilted to the left and to the right from the normal to the smectic layers, one distinguishes 'uniform left' and 'uniform right' structures. If there are two neighbouring 'left' and 'right' domains, the stripes form walls to change orientation by 2α or $\pi - 2\alpha$. In contrast, transitions between a uniform and a twisted state can be arranged without drastic reorientation of the stripes (figure 1).

The most striking feature of the stripes is that their periodicity and orientation strongly depend on the applied voltage (figure 2). Since the optical properties of the stripe textures are modulated, we used optical diffraction to study the field effects, as described below for the uniform state.

A polarized He-Ne laser beam is normal to the cell, and the plane of diffraction is normal to the stripes (figure 4). The diffraction pattern reveals a clear pair of ± 1 st order diffraction maxima and a weak ± 2 nd order diffraction maxima. To measure the time response and diffractive efficiency of the grating, we set the photodetector at the position of the (-1) st maximum. The time of switching of stripe orientation depends on

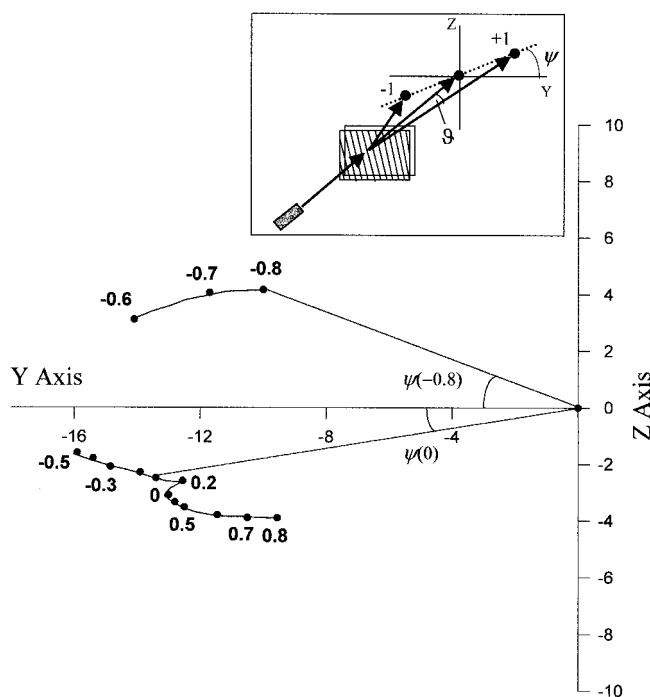


Figure 4. Position of the (-1) -st diffraction maximum as the function of the applied voltage (0.2 Hz square wave signal); $d = 21\ \mu\text{m}$, PVA coating. The inset shows the optical set-up. Z-axis is parallel and Y-axis is normal to the rub direction; both axes are in the plane of the cell.

the applied voltage; the upper limit is about 0.2 s. The measurements were performed at a larger time scale to test the equilibrium states. The diffraction efficiency is weak—1–2%.

As manifested by the shift of the light spots on a screen behind the cell (see figure 4), both polar ϑ and azimuthal ψ angles of diffracted beams change with the voltage. The polar angle ϑ between the incident beam and the diffracted beam depends on the period of modulations, $\sin \vartheta = m\lambda/\Lambda$ (m is the order of the diffraction maximum, λ is the wavelength of the incident light) and the azimuthal angle ψ reflects the in-plane orientation of stripes. The field-induced changes in ϑ and ψ are mostly continuous. However, in a narrow range between (-0.5) and (-0.6) V, both the extinction direction and stripes reorient discontinuously around the rub direction. The reorientation proceeds through the movement of grain boundary walls at which the stripes change their direction. At voltages higher than $U \approx \pm 0.8$ V, the modulations and diffraction disappear.

At $U = 0$ V, one finds $|\psi| = 10^\circ\text{--}15^\circ$; this value is the same as the angle α of stripe orientation (see above) and roughly corresponds to θ_{eff} . At $U \approx \pm 0.8$ V, the azimuthal angle increases to $|\psi| \approx 21^\circ\text{--}23^\circ$, close to the value θ of the director tilt in the SmC* layers.

Complete deciphering of the modulated structure requires detailed X-ray experiments. At this point, we suggest that the stripes are undulations of the (normally straight and horizontal) chevron ridge, figure 5. These undulations relax excessive dilation of smectic layers at the chevron tip.

In smectic phases, variations in the thickness and tilt of layers both contribute to strains. The chevron is basically a tilt of layers needed to compensate the decrease in their thickness. In the SmC* phase, molecular tilt by an angle θ decreases the layer thickness by a factor

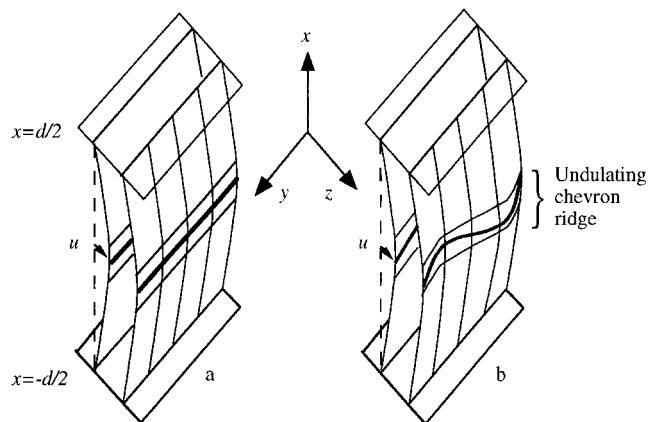


Figure 5. Geometry of smectic layers for a regular chevron (a) and for a chevron with an undulating ridge (b). The periodicity of the undulations is greatly enlarged (as compared with the cell thickness) for clarity.

$1 - \cos \theta \approx \theta^2/2$. This decrease can be compensated by the tilt of layers, which increases the distance between the layers along the z -direction.

The tilt of layers can be expressed through their displacement u along the z -axis as $(u_x^2 + u_y^2)/2$; the subscripts indicate spatial derivatives [7]. Usually (see [4–6] and references therein), it is assumed that θ and u depend only on the coordinate x normal to the cell plates; the chevron ridge is strictly parallel to the horizontal y -axis, figure 5(a). This assumption works very well for thin cells, where the boundary effects restrict the number of configurations. The compressibility term in the free energy density then reads $f = B(u_x^2 - \theta^2)/8$ (B is the compressibility modulus); this term is minimized by adjusting θ and u_x along the x -axis. In the tip region, however, u_x reverses its sign and $u_x \rightarrow 0$ near the middle plane $x = 0$; the strain caused by the molecular tilt is not properly compensated and θ has to be noticeably smaller than its equilibrium bulk value [6].

With $u_x \rightarrow 0$ at the chevron ridge, the strains can still be compensated by the layer tilt in the horizontal yz plane: θ and u depend on both x and y , so that $u_y \neq 0$ and $f = B(u_x^2 + u_y^2 - \theta^2)/8$. Non-zero u_y allows θ to be closer to its equilibrium bulk value. The chevron is periodically undulated (obviously u_y cannot be a constant), figure 5(b). One can visualize this model by tightening a fist: the line of knuckles mimics the configuration of a smectic layer around the chevron tip†. The effect is more plausible in a thick cell and in the deep SmC* phase where the equilibrium θ is large and the tip energy increases [6].

It may be noted that the classic Helfrich–Huraull undulations in smectic and short-pitch cholesteric phases illustrate the very same mechanism of strain compensation by both $u_x \neq 0$ and $u_y \neq 0$ [23, 24]. Parabolic focal-conic domains that sometimes decorate the chevron tips also involve two-dimensional modulations (although the amplitude is much larger than in our case). Recent experiments by Takanishi *et al.* [20] demonstrate spatial oscillations of the chevron ridge in the case of field-induced stripes in thin ($2 \mu\text{m}$) SmC* cells.

In the discussion above we neglected nematic-like contributions, azimuthal direction of the molecular tilt, and effects such as twist deformations. These factors certainly contribute to the electric field effects and to the shaping of the chevron tip which is not a simple planar sinusoidal configuration even when $U = 0$. For example, the fact that the stripes are not parallel to the rub direction suggests that the undulations as seen in projection onto the yz plane are not symmetric: two adjacent shoulders of the undulation wave are of different length. Such asymmetric undulations are known in lyotropic ‘rippled’ phases [25]; they find an explanation

[26] in a model of coupling between the molecular tilt and the shape of bilayers composed of chiral surfactant molecules. Double periodicity that occurs in our samples when the microscope is defocused indicates that the modulations also have a vertical component.

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