Recent advances in nematic and smectic A anchoring on amorphous solid surfaces [1]

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Invited Lecture

Recent advances in nematic and smectic A anchoring on amorphous solid surfaces [1]

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New anchoring properties of liquid crystals on amorphous solid surfaces are presented. In nematics (N), angular anchoring is usually described in terms of the Rapini–Papoular form, assuming constant surface order parameter. We generalize this expression, predicting a decrease of surface order for strong surface disorientation. Recent experiments on anchorings of varying strength confirm these predictions. Conjectures for the angular anchoring of smectic A on a solid amorphous surface explain the two easy layer orientations, normal to the surface or parallel, faceting inside a small critical angle. Roughness-induced surface transitions are discussed. For antagonistic nematic and smectic anchorings, we expect, below the N–S_A transition, a bent nematic surface boundary layer, recently observed by smectization under an electric field. Finally, the positional anchoring strength of smectics is introduced in terms of shear induced surface melting, and confirmed by a recent observation of oscillating shear stresses at the layer period.

1. Introduction

This paper is a short review of some experiments made recently in Orsay, on the anchoring properties of liquid crystals on amorphous solids. We discuss the disorientation induced surface disordering of nematics, the angular anchoring of smectic A layers, when favoured or constrained by a nematic anchoring, and finally the positional anchoring of smectic layers. Some effects are supported by experimental results and can be considered as real, others are somewhat speculative and presented mostly to stimulate further work.

2. Disorientation induced surface disordering in nematics

Angular anchoring of nematics on amorphous planar substrates is traditionally described by the Rapini–Papoular (RP) form [1]

$$W_s = -(1/2)(K/l)(\mathbf{n} \cdot \mathbf{n}_o)^2, \tag{1}$$

where $K(\sim 10^{-6} \text{cgs})$ is the nematic curvature constant, l an extrapolation length ($l \sim 0.1$ to $1 \ \mu m$), $\mathbf{n}$ the nematic director at the surface and $\mathbf{n}_o$ the easy orientation induced by the surface. Let us restrict ourselves in terms of symmetry to $\mathbf{n}_o$ normal to (homeotropic) or inside (planar) the solid surface. The RP form works well for weak anchorings ($l \sim 1 \ \mu m$), but not for stronger ones ($l \sim 0.1 \ \mu m$), where higher power angular terms seem to be involved [2]. We wish to explain this behaviour with a simple model generalizing the RP form.

We assume that the anchoring is produced by a surface quadrupolar field $\sim \mathbf{Q}_o = (3/2)^{1/2} S_o(\mathbf{n}_o \mathbf{n}_o - \mathbf{I}/3)$. S_o is the scalar order parameter favoured from the surface field oriented along $\mathbf{n}_o$, calculable in principle by a microscopic model. $\mathbf{Q}_o$ is assumed to be uniaxial for simplicity, and traceless, i.e. the surface does not change the nematic

density. The surface–nematic interaction free energy can then be represented as

$$W_s \sim \mathrm{Tr}\,(\mathbf{Q}-\mathbf{Q}_o)^2, \tag{2}$$

where $\mathbf{Q}_o = (3/2)^{1/2} S_o (\mathbf{n}_o \mathbf{n}_o - \mathbf{I}/3)$ is the surface quadrupolar order parameter of the nematic. Equation (2) is just a particular case of more elaborated surface energy forms [3]. Let us show that equation (2) predicts simply a decrease of S when the nematic is disoriented at the surface. Expression (2) becomes

$$W_s \sim S^2 + S_o^2 - SS_o(3(\mathbf{n}\cdot\mathbf{n}_o)^2 - 1). \tag{3}$$

S which minimizes W_s is

$$S_m = \frac{S_o}{2}(3\mathbf{n}\cdot\mathbf{n}_o)^2 - 1). \tag{4}$$

For $\mathbf{n}=\mathbf{n}_o$, $S_m = S_o$ as expected. If we could fix $S = S_o$ at the surface, we would keep for W_s the RP form. In fact, this is not possible. When disorienting the surface, S_m should decrease down to $-S_o/2$ for $\mathbf{n}\perp\mathbf{n}_o$. This negative S value has a simple meaning: we force the uniaxial orientation $\mathbf{Q}$ of a nematic perpendicular to the surface field orientation $\mathbf{Q}_o$. In this case, the nematic material will take a discotic orientation so that, at least, half of the surface molecules are well oriented along the surface field (see figure 1).

In practice, to change the surface orientation, we need to use an orienting electric (or magnetic) field, coupled for instance with the dielectric anisotropy of the nematic. The total volume torque density from the applied field can be integrated to give the surface torque which distorts the surface orientation. The surface order S is now calculable from the balance between the bulk nematic action, which tends to force $S = S_B$ (the bulk order parameter), and the surface, which attracts S towards S_o. These two forces are respectively proportional to ξ^{-1} and l^{-1} (ξ is the coherence length of the nematic–isotropic transition). The surface order decrease, from the surface disorientation, is then of the order of ξ/l, in the limit where l is short enough (strong anchoring), but weak compared to ξ. The angular dependence of $S_m \sim (\mathbf{n}\cdot\mathbf{n}_o)^2$ introduces in W_s a term $\sim(\mathbf{n}\cdot\mathbf{n}_o)^4$, which explains the observed higher order terms in W_s previously measured for strong anchorings. The dependence of the disordering of a disoriented surface upon the anchoring strength has been recently shown to vary indeed as ξ/l. The detailed experiment will be published elsewhere [4].

Another experiment could lead to an analogous conclusion. The surface orientation of a bistable oblique anchoring, created by an oblique SiO evaporation, has been
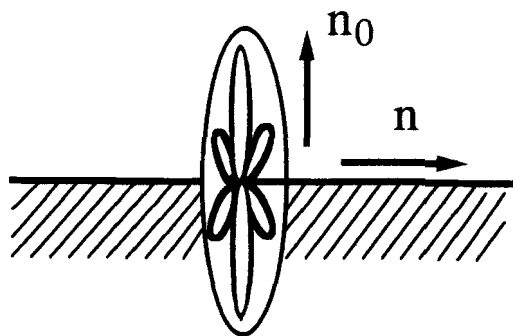

Figure 1.   Discotic ordering of a surface nematic layer, when the director is forced to be normal to the surface field orientation.

measured in the presence of a strong electric field forcing the orientation towards the normal to the surface. We first observe the onset of a surface bifurcation, where the two surface orientations merge. At high enough field, the anchoring seems to become much weaker. This could be another manifestation of a disorientation-induced surface disordering, although another explanation in terms of surface friction cannot be discarded. This experiment will be described elsewhere [5].

Note finally that the disorientation induced surface disordering should not be confused with the permanent decrease of order observed in the absence of (or for a small) disorientation, close to some solid interfaces [6,7], which can be partly attributed to surface roughness [8].

3. Angular anchoring of smectic layers

Smectic A materials are well known to orient, close to an amorphous (glass) solid surface, along two possible easy directions: parallel or perpendicular to the planar substrate. In what follows, we propose a model to explain these preferred orientations. This model is further extrapolated to imagine the possible behaviour of smectics on rough surfaces.

3.1. Anchoring on a flat surface

The large change of density at a smectic–glass interface couples with the smectic order to orient the layers parallel to the solid substrate. To tilt the smectic texture at an angle α from a planar solid surface (see figure 2), we must spend some energy. We assume the macroscopic size L of the sample to be infinitely large, so that we cannot relax elastically the tilt α. To accommodate the tilted texture, we have to create a dislocation per layer on the glass surface. In a continuous description, the dislocation core size compares with ξ, the coherence length of the smectic–nematic transition. Calling m the layer thickness, the distance between dislocations for small α is m/α, i.e. there exists a critical angle $\alpha_c = m/\xi$ for which the dislocations merge and build a melted grain boundary. To estimate the energy of the dislocations and of the grain boundary, we use the standard Landau–de Gennes [9] free energy density, expressed in terms of the smectic order parameter Ψ

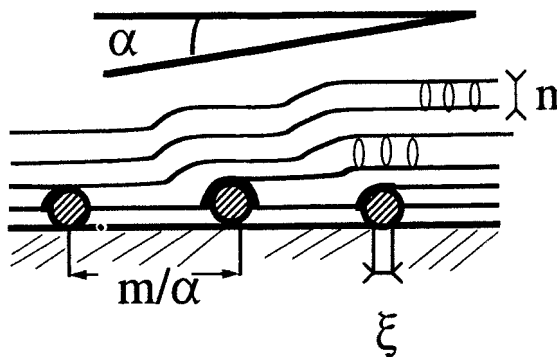$$f = A\Psi^2 + \tfrac{1}{2}B\Psi^4 + C(\nabla\Psi)^2.\tag{5}$$

Figure 2. Tilted smectic facet, with a periodic array of dislocations, for smectic layers almost parallel to a planar solid boundary.

162   G. Durand

As usual, $A = \beta\,(T - T_c)$, where T_c is the transition temperature. At equilibrium, in the uniform bulk, Ψ and ξ are given below T_c by

$$\Psi_B^2 = -A/B, \quad \xi^2 = -C/2A = C/(2B\Psi_B^2) \tag{6}$$

and the minimum condensed bulk energy density is $-A^2/2B$. Per unit length, the dislocation free energy is then of the order: $(-A^2/2B)\xi^2 \sim C\Psi_B^2$. With the dislocation density $|\alpha|/m$, the dislocation network surface free energy is

$$W_{\mathrm{disl.}} \sim C\Psi_B^2|\alpha|/m. \tag{7}$$

On the other hand, for $\alpha \geqslant \alpha_c$, the free energy of the grain boundary is

$$W_{\mathrm{gb}} \sim C\Psi_B^2/\xi \sim (BC)^{1/2}\Psi_B^3 \tag{8}$$

independent of α. In fact, the gradient term C (and then ξ) is anisotropic and should have been written as $C_\parallel \cos^2 \alpha + C_\perp \sin^2 \alpha$. Finally we can represent a plausible angular anchoring energy of smectic layers as sketched in figure 3. The non-analytic surface energy dependence $\sim |\alpha|$ is characteristic of a stable smectic facet, parallel to the glass. The angle α_c corresponds to a roughening transition, above which all tilts can be accommodated by a melted grain boundary. Because of the smectic anisotropy, the layer orientation perpendicular to glass corresponds now to a metastable state of energy $\sim (C_\perp B)^{1/2}|\Psi|_B^3$.

3.2. *Anchoring on a rough surface*

We imagine a sine wave undulating surface with a period $L \gg \xi$, an amplitude a ($a < L$), and a mean tilt $\alpha \sim a/L$. We suppose that the smectic layers far from the surface orient parallel to the mean glass plane orientation. The problem is to estimate how the smectic material can match the surface undulations. A rough surface would be represented by a random superposition of such undulations.

Close to the S_A–N transition, it is probably simpler for the material to fill the surface undulations by melting into the nematic state (see figure 4). The associated surface energy is

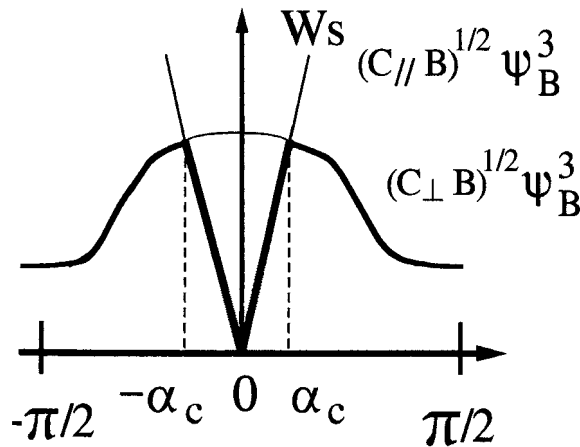$$W_{\mathrm{m}}/\mathrm{cm}^2 \sim B\Psi_B^4 a. \tag{9}$$

Figure 3.   Possible angular dependence of a tilted smectic texture surface energy. α_c defines the roughening transition between the faceting texture of figure 2 for small α, and a melted surface grain boundary texture for large α.
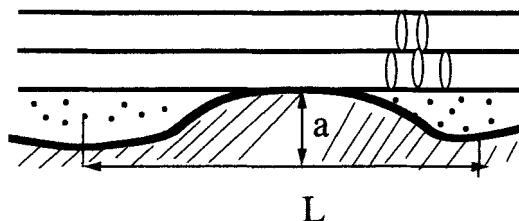
Figure 4. Smectization close to an undulating surface: the filling represents nematic material.

When $\xi < a$, if $a/L > \alpha_c$, we can fill the undulations with parallel layers which produce at the surface a grain boundary of thickness ξ (see figure 5), with energy

$$W_g \sim (BC)^{1/2}|\Psi|^3. \tag{10}$$

For small angles $a/L < \alpha_c$, we can fill the undulations with parallel layers and surface dislocations (see figure 6). The surface energy is

$$W_d \sim C\Psi_B^2 a/(mL). \tag{11}$$

Finally, there must be a temperature where the smectic material fills all the available space, allowing its layers to undulate along the surface without any defect (see figure 7). The undulations propagate normal to the interface on a length [10] $L' \sim L\,(L/\xi) \gg L$. The elastic energy involved is of the order

$$W_e \sim (C/m^2)\Psi^2(\alpha/L')^2 L' \sim Ca^2|\Psi|m^{-1}L^{-2}, \tag{12}$$

where $(C/m^2)\Psi^2$ is the usual layer compressional elastic constant $B_\parallel$.

The free energies in equations (9) to (12) vary in temperature as the powers 4, 3, 2, 1 of $|\Psi| \sim m/\xi$. Cooling down the smectic below T_c, we expect transitions to occur between the corresponding surface textures. W_m and W_g compare for $\xi_{mg} \sim a$. W_m and W_d compare for $\xi_{md} \sim (mL)^{1/2}$. W_m and W_e compare for $\xi_{me} \sim (m^2L^2 a^{-1})^{1/3}$. W_d, W_g and W_e
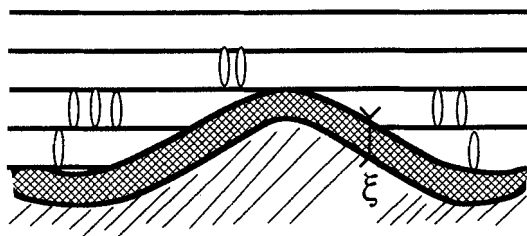

Figure 5. Filling with parallel smectic layers and a surface grain boundary.
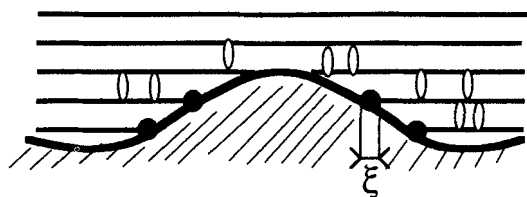

Figure 6. Filling with parallel smectic layers and surface dislocations.

compare for $\xi_{dge} \sim mL/a$. For a rough surface, when $a^2 > mL$, the volume melted texture of figure 4 should occur first close to T_c, followed by the surface melted texture of figure 5 at $\Delta T_{mg}/T \sim (m/a)^2$. A last transition to the undulated texture of figure 7 should occur at $\Delta T_{ge}/T \sim (a/L)^2$. For a smooth surface, when $a^2 < mL$, we should go directly from the volume melted texture to the undulated texture at $\Delta T_{me}/T \sim (ma/L^2)^{2/3}$. For $a^2 = mL$, all these surface transitions should happen at the same temperature. With reasonable values of $m/L \sim (a/L)^2 \sim 10^{-2}$, we find $\Delta T \sim$ a few degrees, i.e. easily observable. These first order surface textural transitions should present some temperature hysteresis. It would be interesting to check if they do really exist.
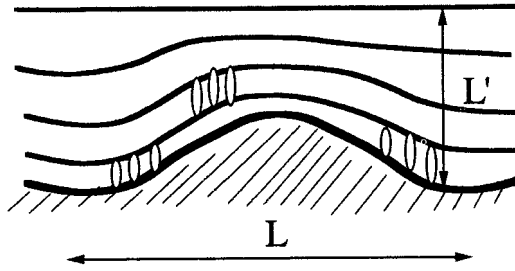

Figure 7. Filling with elastically relaxed undulating layers.

4. Antagonistic nematic and smectic angular anchorings

Up to now, we have not considered the effect of nematic anchoring on the orientation of smectic layers. A simple situation is the one of planar or homeotropic nematic anchorings, which are known to force the smectic layers perpendicular or parallel to the surface. Let us consider a strained situation, where two parallel glass boundaries are treated to favour a nematic homeotropic alignment on one side, and a planar one on the other side (see figure 8). We apply an electric field $\mathbf{E}$, normal to the plates. The liquid crystal dielectric anisotropy $\varepsilon_a > 0$ forces the molecules of the nematic to bend from the planar orientation to the homeotropic, over the electric coherence length ξ_E defined by $K/\xi_E^2 = (\varepsilon_a/4\pi)E^2$. ξ_E is assumed to be smaller than L, the sample thickness. The nematic anchoring strength is supposed to be infinite for the moment. We call $\theta(z)$ the nematic angle $(\mathbf{n}, \mathbf{E})$. We imagine that the bent nematic is cooled down below T_c. We wonder what is the smectic arrangement in the cell? This situation has already been discussed in the absence of a field and leads to textural instabilities [11]. We assume E is strong enough to suppress these instabilities.

The smectic is expected to grow easily from the homeotropic plate, where the field $\mathbf{E}$ and the plate orient the molecules in the right direction. Further away, the curved nematic is tilted by θ from the layer normal. This situation was discussed long ago by de Gennes [6], in his parallel between smectics and superconductors. The smectic tries to expel the curvature field $d\theta/dz = \theta'$ as a superconductor would do for a magnetic field $\mathbf{H}$. The curvature should be damped off inside the smectic, on a length

$$\lambda \sim (K/C_\perp)^{1/2}(m/|\Psi|) \sim K/C_\perp)^{1/2}\xi. \tag{13}$$

For $\lambda < \xi$, the smectic (so-called type I) should resist the curvature θ' until a critical value θ'_c (H_c for superconductors), above which it melts to the nematic. For $\lambda > \xi$, the curvature strained type II smectic will let some defects appear to allow a progressive onset of layer bend above θ'_{c1} (H_{c1} for superconductors) and finally should melt for a higher curvature field θ'_{c2} (H_{c2} for superconductors). θ'_c is calculated by writing that the
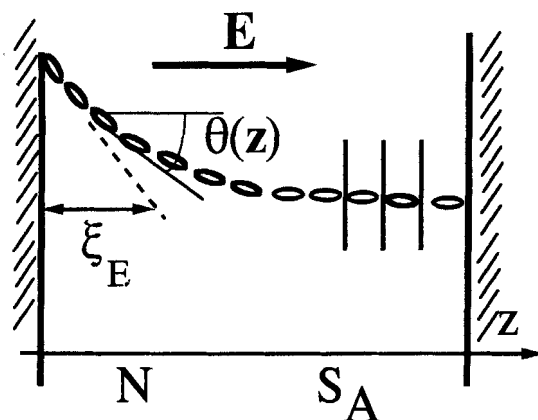
Figure 8. A partly smectized hybrid cell, with a nematic boundary layer in contact with the planar plate where the field **E** concentrates the curvature.

curvature energy equals the smectic condensation energy. With our notations, we get $\theta'_c \sim m/(\lambda\xi)$. As in superconductors, we can estimate $\theta'_{c2} \sim (\lambda/\xi)\theta'_c$ and $\theta'_{c1}(\xi/\lambda)\theta'_c$. Because λ and ξ are expected to be of the same order of magnitude, all critical curvatures should compare.

We can now guess what should happen when cooling the nematic under the electric field. Along the bent nematic profile, the curvature drops continuously from $1/\xi_E$ down to zero. Let us call ΔT_c the temperature shift below T_c for which $1/\xi_E \sim \theta'_c$ or θ'_{c2}, i.e. for which $m\xi_E \sim \xi\lambda$. ΔT_c, defined by $\Delta T_c/T \sim m/\xi_E$, should be easily observable; with a typical $\xi_E \sim 3000$ Å, we expect $\Delta T_c \sim 3°C$. For $\Delta T < \Delta T_c$, the type I or type II smectic feels a varying curvature field which exceeds the corresponding critical value: a nematic boundary layer must exist close to the planar plate. The width d of this nematic boundary layer is defined by writing that the nematic curvature at the nematic–smectic interface is θ'_c or θ'_{c2}. For a weak field, this leads simply to $d \sim \xi\lambda/m \sim mT/\Delta T$, in the μm range for $\Delta T \sim 1°C$. d is macroscopic, because we impose a large angular (flux) anchoring constraint $\theta = \pi/2$ on the planar plate.

For $\Delta T > \Delta T_c$, the behaviour of the smectic should depend on its nature. In a type I material, the nematic boundary layer should persist, since θ remains weak at the interface. d, and accordingly the birefringence, should decrease when cooling, until eventually the nematic anchoring breaks for $l = d$. For a type II smectic, on the other hand, the nematic boundary layer should disappear for $\Delta T \sim \Delta T_c$, and be replaced by a bent Shubnikov smectic texture containing edge dislocations or other defects. The type II birefringence decrease is expected to saturate.

In the limit of a very strong type II material $(\lambda \gg \xi)$, the Shubnikov texture would adjust exactly at all temperatures to the initial curved nematic profile, and no change of birefringence is expected. A more standard type II smectic should give a decrease in birefringence smaller than the one of type I (and a saturation at $\Delta T \sim \Delta T_c$), since the Shubnikov texture is generally more rigid to bend than the nematic.

An experiment has been made in Orsay to check these ideas. The smectic A material CBOOA (4-n-octyloxy-N-(4-cyanobenzylidene)aniline) was used; this compound has a large $\varepsilon_a \sim 10$, and a second order nematic–smectic A transition at $T_c = 83.2°C$. The birefringence of the cell is measured when cooling down through T_c. We do observe a decrease in birefringence, which fits with the decrease in the nematic boundary layer

thickness d. A few degrees below T_c, the birefringence saturates and a hysteretic behaviour is observed. This corresponds to the total smectization of the cell, i.e. indicates that CBOOA is a type II smectic. A report of this experiment will be published elsewhere [12].

5. Positional anchoring strength of smectics A

Up to now, we have assumed that, when smectic layers are obliquely in contact with a planar glass surface, the smectic ordering at the surface is zero. This assumption is based on the fact that the glass is amorphous, and cannot induce a periodic ordering. This is not exactly true: as for nematics, the smectic bulk ordering Ψ_B can force a weak but non-zero surface ordering Ψ_s. For a normal layer orientation, a likely profile of $\Psi(z)$ is sketched in figure 9 (a), where the extrapolated Ψ is zero inside the glass. It probably takes some time to establish the surface ordering Ψ_s, since the random forces which fix the molecules on the glass are relatively large, resulting in a high surface viscosity. The existence of a surface ordered smectic texture is probably the origin of the usually observed surface memory effects like epitaxy, surface induced tilt (when for instance the smectic layer spacing changes with temperature, or at a smectic A to tilted smectic C transition), and so on. Here, we are more interested to estimate the positional anchoring strength associated with this surface smectic texture Ψ_s.

We assume that, by some mechanical action, we move the layers by a quantity x parallel to the surface and normal to the layers. If this motion is rapid enough, the surface smectic texture cannot glide. Each time the lateral motion $x = p.m$ (p is an integer), the displaced layers can match exactly the surface density modulation and no force is needed to keep the layers in place. In between, on the contrary, the equilibrium texture costs some free energy which oscillates then as $(1-\cos kx)$ ($k = 2\pi/m$). The maximum energy corresponds to $x = m/2$. In this situation, the order profile $\Psi(z)$ must be zero above the glass, in the smectic material, since $\Psi(z)$ must be linked continuously to the apparently out of phase (i.e. negative) surface value $-\Psi_s$ (see figure 9 (b)). The energy which breaks the positional smectic ordering is that necessary to displace the melted point $\Psi(z) = 0$ from below to above the surface. In a continuum description, this displacement is of the order of: $z \sim \xi|\Psi_s/\Psi_B|$. The associated surface energy is $(CB)^{1/2}\Psi_b^3 z/\xi \sim (CB)^{1/2}\Psi_b^2|\Psi_s|$, i.e. the positional anchoring energy of a smectic A in the planar geometry should be written as

$$W_p = (1-\cos kx)(CB)^{1/2}\Psi_B^2|\Psi_s|. \tag{14}$$

As Ψ_s is supposed to be proportional to Ψ_B, this energy varies as $|\Psi_B|^3$. Far inside the smectic phase, its order of magnitude is $(k_B T/m^2)|\Psi_s|$. The anchoring of smectic layers at a sheared planar interface is then described as a periodic sequence of surface melting and recrystallization. This one-dimensional melting mechanism could be considered as a model system for solid friction.

An experiment has been made recently in Orsay, to check these ideas. The smectic A material, 4O.8 (4-n-octyl-N-(4-n-butyloxybenzylidene)aniline), in a planar texture of thickness L, is sheared by piezoelectric ceramics which impose a sawtooth, time-varying displacement to one plate. The shear stress σ transmitted across the other plate is measured as a charge appearing across other ceramics. As for plastic crystals [13], we find a periodic oscillation of σ at a period 28 ± 1 Å $\sim m$, the layer thickness, when the shear is normal to the layers, and no oscillation when it is parallel. The measured amplitude of the shear oscillation energy is found, depending on the temperature, to be in the 10^{-5}–10^{-6} erg cm^{-2} range, which results in $\Psi_s \sim 10^{-2}$–10^{-3}. This oscillation is
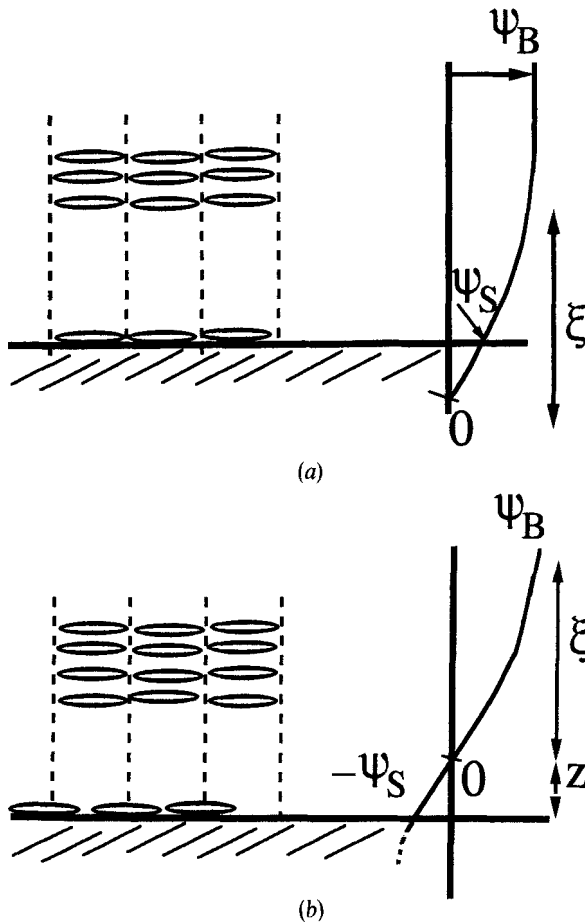
Figure 9.   Variation of the smectic order close to an amorphous surface. (*a*) In absence of shear; the $\Psi = 0$ point is virtual. (*b*) For a half-layer shear; the $\Psi = 0$ point must appear inside the smectic material, increasing the surface energy. The associated periodic melting induced by a uniform shear across the layers defines the smectic positional anchoring energy.

superimposed on a visco-elastic relaxation, with characteristic time 10^{-3} s. This relaxation can be explained from the elastic tilt of the sheared layers in the bulk of the thickness L, with an elastic constant $B_{\perp} = C_{\perp} k^2 \Psi_B^2 \sim 10^8$ cgs, and a viscous flow (of viscosity $\eta \sim 1$ poise) localized close to the surface, in a thickness ξ. This results in a relaxation frequency $(B_{\perp}/\eta)(\xi/L)$, in the kHz range. The shear flow seems to be concentrated close to the boundary solid plates because the lower surface smectic ordering allows locally an easier permeation. A full report of this experiment will be presented elsewhere [14].

6.   Conclusion

We can now give some indications as to that could be interesting to investigate in the future, in the field of liquid crystal anchoring.

In nematics, too much work has been invested on the angular anchoring properties, and too little on the surface ordering. The experiments presented above are the first to

demonstrate the existence of surface disordering related to surface disorientation. Direct measurements of the surface energy dependence on the order parameter should be designed and performed to understand the wetting of strained nematics. In all cases, it will be more fruitful to build Landau-like surface energies, by using the surface nematic order tensor to generalize the Rapini–Papoular form.

In smectic A materials, the experimental situation is far less developed. We have presented the first evidence of the positional anchoring of smectic layers, normal to a sheared interface. When nematic and smectic angular anchorings are antagonistic, a simple extension from the old de Gennes parallel between smectics and superconductors leads to the prediction of a quasi-macroscopic nematic boundary layer, with a different temperature behaviour for type I or II materials. This nematic boundary layer has probably been observed, but further experiments are still needed to confirm its existence. All these static properties of constrained smectics close to solid boundaries are important in order to understand their dynamical behaviour. The simplest flows in smectics, for instance, are far from being understood. The observed concentration of permeative flow close to a boundary, where smectic ordering vanishes, is just an example of such unexpected behaviour.

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