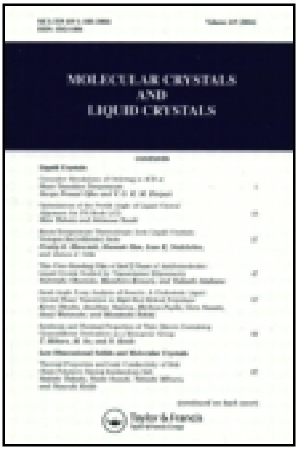

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Anticlinic Surfaces in Freely Suspended Smectic Films Stabilized by Electrostatic Interactions

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Anticlinic Surfaces in Freely Suspended Smectic Films Stabilized by Electrostatic Interactions

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Electrostatic dipolar interactions between transversal polarizations of the finite and parallel surface smectic layers are shown to stabilize the anticlinic smectic-C surfaces in freely suspended smectic films formed from (anti)ferroelectric liquid crystals.

Keywords: antiferroelectric liquid crystals; freely suspended films; dipolar electrostatic interactions

INTRODUCTION

Freely suspended films of smectic liquid crystals are very suitable for observations and measurements. They provide a fine example for physical systems, where the influences of the broken discrete translational symmetry, free surfaces and limited number of smectic layers in film can be studied and the information about the mechanisms, governing the behavior of smectics, can be obtained. Just an agreement of the results of the ellipsometric [1] and resonant X-ray scattering [2] experiments, done on freely suspended films, with the predictions of the discrete phenomenological model for antiferroelectric liquid crystals [3], confirmed at last the suggested structure for the SmC$_\alpha$ phase.

In this contribution we shall concentrate on the anticlinic smectic-C surfaces in freely suspended SmA films. Molecules in the upper and the bottom surface layers are tilted in opposite directions (anticlinic), while in the interior layers of the film the structure corresponds to the SmA phase, see Figure 1. If the molecules in the upper and the bottom surface layers are tilted in the same direction, the structure is called the synclinic smectic-C surfaces. Where the molecules are tilted also the induced transversal polarization exists, with the magnitude proportional to the magnitude of the tilt, parallel to the layers and perpendicular to the tilt vector. The upper and the bottom ferroelectric polarizations are antiparallel in the anticlinic smectic-C surfaces, and parallel in synclinic smectic-C surfaces. Un-

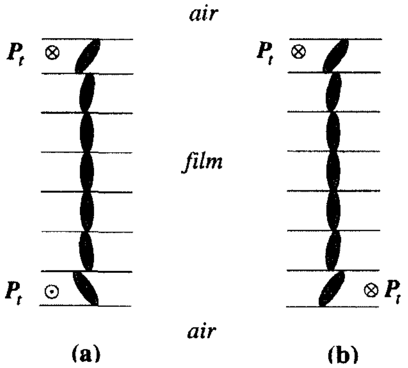

Figure 1: A schematic representation of the anticlinic (a) and synclinic (b) smectic-C surfaces. The magnitudes of the tilt and polarization are large in the surface layers and small in the rest of the film. For the anticlinic smectic-C surfaces the polarizations of the upper and the bottom surface layers are antiparallel and for the synclinic smectic-C surfaces parallel.

expectedly the anticlinic smectic-C surfaces were found as a ground state in freely suspended films of some smectic liquid crystals with (and without) anticlinic bulk phases [4].

As we will show elsewhere [5], ferroelectric liquid crystals, which have only synclinic bulk phases, may in freely suspended films form also partially anticlinic structures. This phenomenon is due to the presence of opposite and frustrating interactions between the nearest and next-nearest layers, as viewed within the discrete model, but the frustration not being strong enough to cause the appearance of anticlinic structure in bulk. With a proper choice of the model parameters together with a consideration of vigorously enhanced order in surface layers, which strengthen also the frustration effect in few surface layers, we are able to find either synclinic or anticlinic smectic-C surfaces as the ground state structures in films. However the difference in the energies of both, as calculated within the simple model, may sometimes not be large enough to account for the measured critical electrical fields, that induce the transition between them.

Therefore in this contribution we consider additionally the electrostatic coupling between the transversal ferroelectric polarizations of the two surface layers. We emphasize that the same kind of interaction between the other layers can be neglected, since the tilt and consequently the magnitude of the ferroelectric polarization is substantial only in the surface layers. It was shown that the two *infinite*, non-correlated, uniformly polarized liquid smectic planes do not interact electrostatically [6]. Although the size of the real freely suspended film samples, or domains separated by domain walls, is indeed macroscopic, but is not infinite. We shall calculate the electrostatic dipolar interaction energy of the two *finite* parallel surface layers, each uniformly polarized. We will show that the dipolar interaction energy decreases with the size of the layers, but rather slowly, and should therefore not be neglected. The electrostatically preferred relative orientation of the tilt in upper and the bottom layers is shown to be anticlinic.

DIPOLAR INTERACTIONS BETWEEN FINITE LAYERS

We shall calculate the energy of the dipolar electrostatic interaction between the upper and the bottom surface layers in freely suspended film. There are $n+1$ parallel smectic layers in the film, the thickness of individual layer is d_0. Film is square in shape and of dimension $L \times L$. Each surface layer is assumed to be uniformly polarized, with the polarization parallel to the smectic plane and perpendicular to the tilt.

Electrostatic interaction between two dipoles $\boldsymbol{p}_1$ and $\boldsymbol{p}_{n+1}$ from

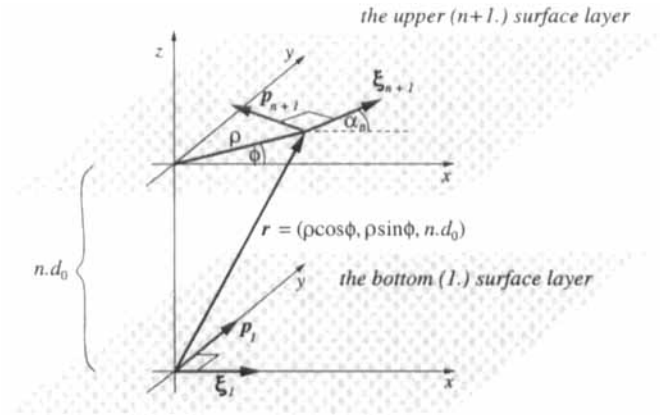

Figure 2: Geometry. Smectic planes are perpendicular to the z axis. Tilt vectors $\boldsymbol{\xi}_1$ and $\boldsymbol{\xi}_{n+1}$ lie within the smectic plane, α_n is the angle between them. Ferroelectric dipoles $\boldsymbol{p}_1$ and $\boldsymbol{p}_{n+1}$ also lie within the smectic plane and are perpendicular to the tilt vectors.

the first and the $(n+1)$-th layers is

$$V(\alpha_n, n, \rho, \varphi) = \frac{|\boldsymbol{p}_1| \cdot |\boldsymbol{p}_{n+1}|}{4\pi\varepsilon\varepsilon_0} \, v(\alpha_n, n, \rho, \varphi) \,, \qquad (1)$$

$$v(\alpha_n, n, \rho, \varphi) = \frac{\left[(nd_0)^2 - \frac{1}{2}\rho^2\right] \cos\alpha_n + \frac{3}{2}\rho^2 \cos(2\varphi - \alpha_n)}{\left[(nd_0)^2 + \rho^2\right]^{5/2}} \,, \qquad (2)$$

where ε is a static dielectric constant of the liquid crystal, nd_0 is a distance between the layers, $\boldsymbol{r} = (\rho\cos\varphi, \rho\sin\varphi, nd_0)$ is a vector between the points, where the two dipoles are situated (see Figure 2) and α_n denotes the relative orientation of the dipoles.

Energetically favorable relative orientation of two dipoles from separate layers depends on ρ [7],

$$\rho < nd_0\sqrt{2} \implies \alpha_n = \pi \,,$$
$$\rho > nd_0\sqrt{2} \implies \alpha_n = 0 \,.$$

Two dipoles close to one above the other prefer to be antiparallel and those farther want to be parallel.

Dipolar interaction between two uniformly polarized parallel surface layers is, from Eq.(1),

$$V_{1,n+1}(\alpha_n) = \frac{|\boldsymbol{P}_1| \cdot |\boldsymbol{P}_{n+1}| d_0^2}{4\pi\varepsilon\varepsilon_0} \int_{S_1} \int_{S_{n+1}} v(\alpha_n, n, \rho, \varphi) \, dS_1 dS_{n+1} \,. \qquad (3)$$

Infinite (S_1, $S_{n+1} \to \infty$) non-correlated uniformly polarized layers do not interact electrostatically [6]. Positional correlations of molecules from neighboring layers imply non-zero dipolar interactions extending up to the next-nearest neighboring layers [7].

For finite and non-correlated square layers, uniformly polarized, one above another and $S_1 = S_{n+1}$, we have calculated numerically the difference between the dipolar interaction energy for parallel orientation of the upper and the bottom surface polarizations ($\alpha_n = 0$) and dipolar interaction energy for antiparallel orientation ($\alpha_n = \pi$),

$$\Delta V_{1,n+1} = V_{1,n+1}(\alpha_n = 0) - V_{1,n+1}(\alpha_n = \pi) \,, \qquad (4)$$

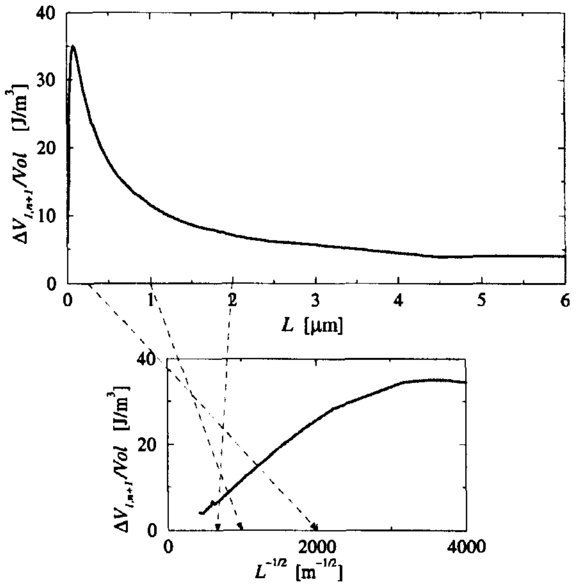

Figure 3: Results of numerical calculations. Difference in dipolar interaction energies $\Delta V_{1,n+1}$ per unit volume of parallel ($\alpha_n = 0$) and antiparallel ($\alpha_n = \pi$) orientations of polarizations $\boldsymbol{P}_1$ and $\boldsymbol{P}_{n+1}$ of the bottom and the upper surface layers. The results are burdened with numerical error, that is less then 1% for $L < 1\mu$m and 10% for $L > 3\mu$m.

for $n = 9$ (film with ten smectic layers), in dependence of the size $S_1 = S_{n+1} = L \times L$ of the square layer. We have not considered the screening effects. The results of numerical calculations are presented in Figure 3. For the material parameters that we have used in our calculations we have chosen the typical values; smectic layer thickness $d_0 = 3$ nm, polarization of the surface layers $P = 40$ nC/cm^2 and static dielectric constant $\varepsilon = 4$ [8]. Only the upper and the bottom surface layers posses transversal polarizations, all the rest are not polarized.

From the Figure 3 we can observe, that the dipolar interactions between two surface layers favor antiparallel orientation of the surface polarizations, thus anticlinic smectic-C surfaces, since $\Delta V_{1,n+1}$ is everywhere positive (see Eq. (4)). The energy difference $\Delta V_{1,n+1}$ does decrease with increasing size of the sample, yet rather slowly and has macroscopic values also for large samples. An estimation for the size, that would correspond to the real sample or domain, is 100 μm [4]. This is one order of magnitude larger than the largest dimension that we have taken and can not be handled numerically in reasonable time. However, from the bottom figure we may guess, that the size dependence of the $\Delta V_{1,n+1}$ is somewhere near

$$\Delta V_{1,n+1} \propto L^{-1/2}$$

for large L. Although this may not be the precise dependence, $\Delta V_{1,n+1}$ obviously decreases even slower that $1/L$. For a hundred microns large domain the interaction energy should be near 1 J/m^3.

For a comparison let us calculate the energy density of the polarized surface layer in an external electric field, parallel to the layers. For the polarization of the layer we take the same value as before (40 nC/cm^2) and for the electric field the approximate value of the critical field, that drives the transition between the anticlinic and the synclinic smectic-C surfaces [4] (5kV/m). The estimated energy density is 2 J/m^3. We see, that the calculated energy difference $\Delta V_{1,n+1}$ is comparable to the energy of the polarized layer in external electric field.

CONCLUSIONS

Dipolar electrostatic interactions between two uniformly polarized, finite surface layers prefer antiparallel polarizations of the surface layers and anticlinic smectic-C surfaces. The upper limit of the energy difference between synclinic and anticlinic orientation, due to dipolar interactions, is macroscopic.

Contribution of electrostatic dipolar interactions between the surface layers to the free energy should probably not be neglected, at least at temperatures, where the tilt and consequently polarization are substantial only at surfaces.

The results obtained also lead us to think, that interactions between separate layers in general may be dependent on the size of the layers more than we thought. And they are maybe important and extend not only between the nearest and the next-nearest layers, but also between more distant layers.

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