Mol. Cryst. Liq. Cryst., 1987, Vol. 151, pp. 417-424. Photocopying permitted by license only. © 1987 Gordon and Breach Science Publishers S.A., Printed in the United States of America.

## FILLING OF SPACE BY FLEXIBLE SMECTIC LAYERS

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Abstract A model of filling of space by smectic A layers is proposed. For large scales r>r the filling is produced by an iterative system of focal conic domains of consequently decreasing sizes. The size of the base of the smallest domains is r. This size is governed by geometry of system, splay elastic constant, and surface tension anisotropy. For r<ra>r</a> all the remaining interstices are filled with layers of spherical curvature.

Quite a number of physical systems, the simplest of which is a smectic A liquid crystal Sm A, possess structures consisting of equidistant fle-xible layers. If Sm A is located within a bounded volume, then the layer distribution is governed not only by the constant thickness condition, but also by the boundary conditions. In the general case, these conditions are satisfied simultaneously due to the origination of defects: walls, lines, and points. However, observations of Sm A textures reveal only lines and points. The latter are associated with two classes of equidistant surfaces, namely, the Dupin's cyclides and the concentric spheres. The cyclides lie within cones of revolution which are referred to as focal conic domains.

Though the history of the problem is long, no final understanding is still achieved of how the

domains shaped as revolution cones and, perhaps, as spheres can continuously fill the space.

As it was shown by W.L.Bragg<sup>1</sup>, for realistic samples this problem reduces to the problem of filling the piramids. According to the basic iterative filling model of R.Bidaux et al.<sup>2</sup>, in order to fill a volume of piramid, one begins with one cone of maximum size, then fills the remaining interstices with smaller cones, etc., down to the molecular length. In the second model, proposed by J.P.Sethna and M.Kléman<sup>3</sup>, a smooth transition between cones is provided by a system of layers of spherical curvature centered on the common apex of domains and the piramid.

In this contribution, we give account of the results obtained in terms of our recent model.<sup>4,5</sup>
The filling patterns are shown to be different for scales smaller and greater than some critical scale ro.

For large scales L>r>ro, where L is the characteristic size of the system, the filling is produced by an iterative system of focal conic domains of consequently decreasing sizes. The size of the base of the smallest domains is ro. For smaller scales  $r < r_0$  we introduce, instead of the focal conic domains hierarchy, new type of packing: all the remaining interstices are filled with layers of spherical curvature.

To comprehend qualitatively the physical nature of the filling pattern proposed, it is sufficient to point out two factors whose competiti-

on determines the equilibrium value of  $r_0$ . The first factor is the difference of elastic energies of the two types of packing. The elastic energy per unit volume is determined by the values of the main radii of layer curvatures:  $g\sim(1/R_1+1/R_2)^2$ . For spherical curved layers  $R_1=R_2>0$ , whereas  $R_1$  and  $R_2$  have different signs for Dupin's cyclides and thus the elastic energy can be reduced. However, this reduction may be prevented by the second factor, namely, the difference in the surface energies of regions with spherical packing (molecules are either normal or incline to the basal plane) and focal conic packing (molecules lie in the basal plane), see Fig. 1. Minimizing the

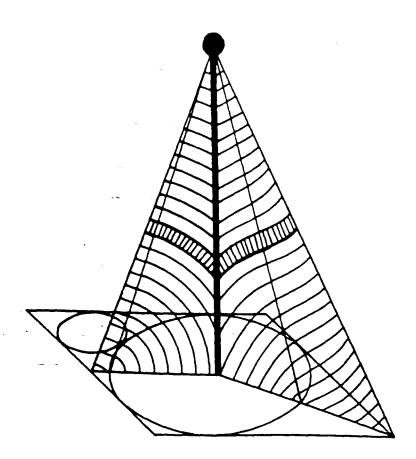


FIGURE 1. Distribution of layers within piramid

sum of elastic and surface energies yields the equilibrium value of  $\mathbf{r}_{o}$ 

$$r_0 = \frac{aL}{b + \Delta 6 L/K}$$
 (1)

where a and b are numerical constants governed by the geometry of the system (~1-10), K is the splay elastic constant, and  $\Delta \mathcal{F} = \mathcal{F}_1 - \mathcal{F}_{11}$  is the surface tension anisotropy:  $\mathcal{F}_L$  is the surface tension at the boundaries of regions with spherical packing,  $\mathcal{F}_{11}$  is the surface tension at the boundaries of regions filled by Dupin's cyclides.

The basic feature of model, the macroscopic character of  $r_0$ , is shown experimentally by numerous Sm A textures. For example, the polarizing microscope photographs in Fig.2 unquestionably show the large clear gaps between the bases of focal conic domains (Fig.2a:C<sub>8</sub>H<sub>17</sub>O-Ø-C<sub>2</sub>N=CH-Ø-C<sub>7</sub>H<sub>15</sub>

t=55°C; Fig.2b: CF<sub>3</sub>O-Ø-CN=N-Ø-C<sub>4</sub>H<sub>9</sub>, t=45°C). These gaps are filled with spherical layers, as it follows from the optical patterns.

As L increases,  $r_0$  asymptotically tends towards a constant value aK/ $\Delta G$ , and this is confirmed by experiment, see Fig. 3. Also we find that  $r_0$  increases by more than 10 times at phase transition Sm A - Sm B, due to the increasing of K in accordance with (1), Fig. 4.

As follows from (1), the filling pattern is very sensitive to the surface tension anisotropy. If  $\Delta 6 < -bK/L$ , then no iterative system of focal conic domains appears. For  $\Delta 6 > -bK/L$ , an ener-

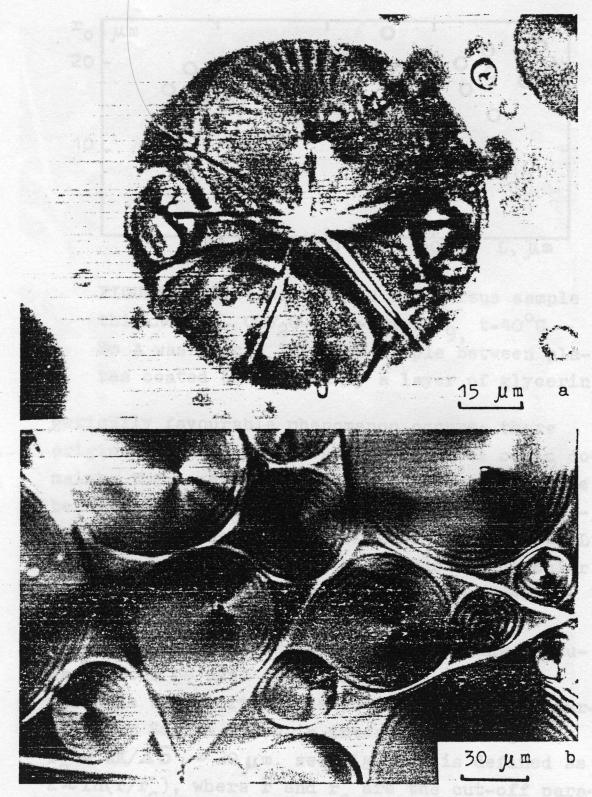


FIGURE 2. Sm A textures: spherical drop freely suspended in glycerin-lecithin matrix (a) and flat capillary coated by indicated matrix

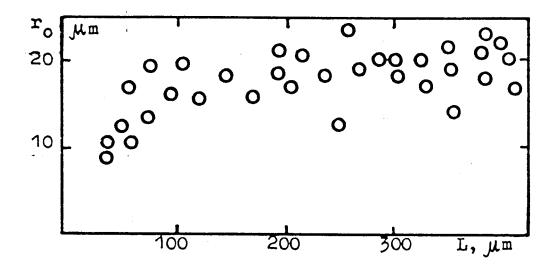
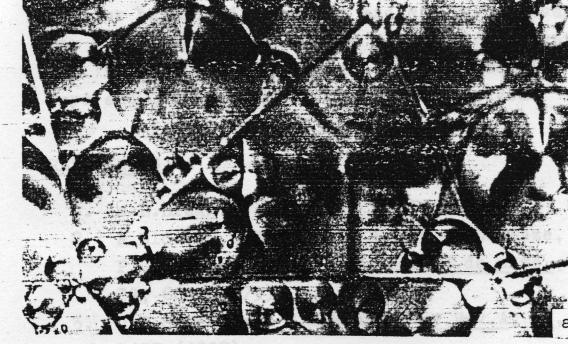


FIGURE 3. Critical radius  $r_0$ , versus sample thickness L.  $CHF_2O-\emptyset-CH=N-\emptyset-C_4H_9$ ,  $t=40^{\circ}C$ . Sm A was placed in wedge sample between plates coated beforehand by a layer of glycerin.

getically favourable phenomenon occurs: there originates an iterative system of focal conic domains, the base radius of the smallest of these being equal to  $r_0$ . As  $\Delta \delta$  increases, the critical radius consequently decreases from  $r_0^{\text{max}} \sim L$  to  $r_0^{\text{min}}$  of the order of magnitude of molecular length. The latter limiting case reproduces the results of Ref.<sup>2</sup>.

If the parameters of model are known, formula (1) can be used to estimate the  $\Delta \delta$  value, which is an important characteristic of Sm A surface. For Sm A - glycerin interface at large L,  $r_0 = aK/\Delta\delta \sim 20 \,\mu\text{m}$ , see Fig.3. a is defined as  $2\pi \ln(\bar{r}/r_c)$ , where  $\bar{r}$  and  $r_c$  are the cut-off parameters for focal conic domains. If  $K \sim 10^{-11}$  N, a-20, we would have  $\Delta \delta \sim 10^{-5}$  N/m, at Sm A - glycerin interface.



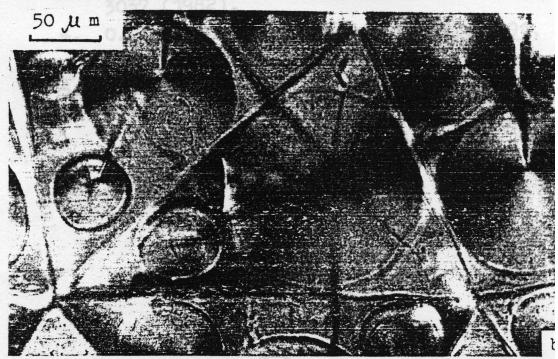


FIGURE 4. Disappearance of small focal conic domains during Sm A - Sm B transition.

CHF 0-Ø-CH=N-Ø-C4H9, t=40°C (a), t=20°C (b).

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