Hierarchy of defect structures in space filling by flexible smectic-A layers

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The question of the filling of space by flexible layers of constant thickness is considered for the example of smectic-A liquid crystals. The possibility of space filling by domains of two types (confocal and spherical) is proved experimentally. A general space-filling model is proposed, according to which, on scales exceeding a certain critical size ρ^*, the space filling is realized by an iterative system of confocal domains (the radius of the base of the smallest domain is approximately equal to ρ^*), and on scales smaller than ρ^* the remaining gaps are filled by spherical layers. An expression is obtained for the critical scale ρ^*, which depends on the characteristic size of the system, the bending modulus of the layers, and also the anisotropy of the surface tension at the boundary of the liquid crystal.

1. INTRODUCTION

A great many physical systems, of which the simplest is a smectic-A liquid crystal (SALC), possess a structure consisting of flexible layers of constant thickness. Each layer of an SALC consists of rodlike molecules oriented in the direction normal to the layer. The layer thickness a is close to the length of the molecule ($a \sim 10^{-9}$ m), and to change it usually requires energies considerably greater than the energy of bending deformations of the layers. The maintenance of a constant distance between the layers implies that the field lines of the normal n to the layers are everywhere straight lines; in other words, the conditions

$$\mathbf{n} \cdot curl\,\mathbf{n} = 0, \quad \mathbf{n} \times curl\,\mathbf{n} = 0 \tag{1}$$

are fulfilled.

If the layer system is situated in a bounded volume, the distribution of n should satisfy not only the conditions (1) but also conditions on the boundary. Simultaneous fulfillment of these conditions in the general case can be ensured only by the appearance in the system of a certain number of defects—singularities of the field n. In principle, in a three-dimensional medium such defects can be two-dimensional (walls), one-dimensional (lines), and zero-dimensional (points). It is obvious (and this is confirmed both by calculations and experimentally) that line and point singularities are more favored energetically. To these singularities there correspond only two classes of equidistant surfaces—namely, Dupin cyclides and concentric spheres (the latter can be considered as a particular Dupin cyclide). The first to show this was Maxwell in 1868, when solving the problem (similar in many respects to that considered here) of the propagation of wave surfaces when rectilinear wave rays pass through an isotropic medium.[1]

In an SALC, which is an optically uniaxial medium, the distribution of the field n and the defects in it are comparatively easily displayed by means of polarization-microscope studies, since the optical axis coincides with the direction of n. As was established by Friedel and Grandjean[2] on the basis of such observations, for planar SALC samples the most characteristic texture is a so-called texture of confocal domains, in which line defects of regular shape, in the form of ellipses and hyperbolas, are clearly distinguishable. Associated with each pair of lines is a distinct confocal domain, in the form of a cone of revolution whose base is the ellipse and whose apex lies on the hyperbola. The layers within a domain have the shape of Dupin cyclides and are everywhere perpendicular to the straight lines joining any point of the hyperbola to any point of the ellipse (Fig. 1). Usually one observes a whole family of domains that have a common apex and are contiguous along generators. Despite the long history of the question, until now it has not been finally clarified how a system of domains in the form of cones of revolution ensures continuous filling of space and how it is possible for layers to cross smoothly from domain to domain in a manner that does not require the introduction of defects with dimensionality higher than unity into the system.

Two theoretical models are known. In the Bragg model of iterative space filling,[3,4] the gaps between large domains are occupied by smaller domains, and so on down to molecular scales (Fig. 2). However, the situation in a real experiment is not always like this, and between domains it is possible that gaps free from defects can be preserved. In the second model, recently proposed by Sethna and Kléman,[5] a smooth transition between domains within one family is realized by a system of spherical layers. However, there is no adequate experimental confirmation of this model either.

In the present paper, on the basis of an experimental study of SALC textures in different geometries, we propose a general space-filling model that is, in essence, a combination of the two previously known models. It is shown that the pattern of the space filling is different for scales greater and smaller than a certain critical size. The dependence of this size on the parameters of the system is found.

2. FILLING OF SPHERICAL VOLUMES

We consider first of all the character of the filling of the simplest model object—a spherical SALC drop freely suspended in an isotropic matrix. The choice of such an object makes it possible to investigate in detail the dependence of
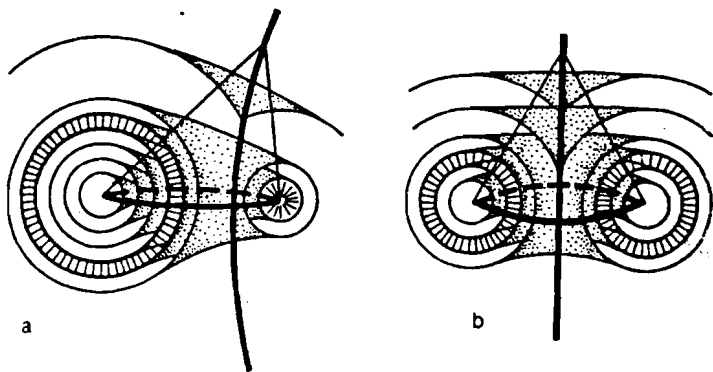
FIG. 1. Smectic-A layers in the form of Dupin cyclides with line defects in the form of a confocal ellipse and hyperbola (a) and a circle-straight-line pair (b). The part of the volume occupied by cyclides and bounded by a surface in the form of a cone of revolution is an isolated confocal domain, whose base is the ellipse (circle) and whose apex lies on the hyperbola (straight line).

the pattern of the space filling on the orientation of the layers at the surface; as will be shown below, the principal features of the filling of a spherical drop are preserved for other experimental geometries too.

We investigated several liquid crystals, but because of their qualitative similarity all the results given in this section pertain just to 4-n-octyloxy-4-n-heptyl-α-cyanostilbene, which possesses a smectic-A phase in the temperature range 52–63 °C. The liquid crystal was dispersed in the form of drops of diameter of the order of 0.1 mm in three isotropic matrices: glycerine, which gives rise to tangential boundary conditions for n (the matrix M_0); glycerine with excess lecithin solution (10 wt%), which gives rise to normal boundary conditions (the matrix M_{10}); and, finally, glycerine with a moderate (1%) content of lecithin solution (the matrix M_1). The orienting properties of the matrix M_1 were described earlier in Ref. 6 and differ from those of the matrices M_0 and M_{10} in the comparatively low value of the anisotropy of the surface tension.

In the indicated matrices, respectively, three types of structures were observed in the drops: I) disordered structures with a large number of defects and bends, II) strongly radial structures with only one defect (a point "hedgehog") at the center of the drop,[7] and finally, III) intermediate structures containing both a point defect at the center and a certain number of line defects, namely, circles on the surface of the drop and straight lines passing through the centers of these circles and the center of the drop (Fig. 3). It is the intermediate structures III that are of greatest interest for discussion.

In the structure of type III two types of regions, with different packings of the layers, can be clearly distinguished. The behavior of the textures under observation in crossed Nicols, and also upon introduction of a quartz wedge, makes it possible to conclude that regions of the first type have a spherical packing of layers, while regions of the second type, in the form of circular cones of revolution, contain layers with the shape of Dupin cyclides and are confocal domains of toroidal configuration (Fig. 1b), built into a ball.

Apart from the indicated pair of lines and the point at the center, there are no other defects in drops with a structure of the type III; the layers pass smoothly between the confocal domains and the regions with spherical packing (Fig. 3b). This is understandable, since the layers intersect the conical surface of contiguity at a right angle. In fact, the

experimental results prove that it is possible in principle to have smooth filling of the gaps between the confocal domains by spherical layers with center of curvature at the point at which the apices of the domains meet.

As a rule, drops in the matrix M_1 contain not two confocal domains, as in Figs. 3a and 3b, but a whole system of contiguous domains (Fig. 3c), the radii ρ of the bases of which are unequal and vary from $\rho \lesssim R$ (R is the drop radius) to $\rho = \rho^* \simeq R/10$. Here the smallest domains are found to be built into the gaps between the larger domains, while the remaining gaps, of sizes smaller than ρ^*, are filled by a single system of spherical layers. We shall consider the physical nature of this filling.

For a qualitative understanding it is sufficient to point to two factors, the competition between which determines the equilibrium value of ρ^* [at a constant temperature, this quantity (for drops) remains unchanged for many hours]. The first factor is the difference in the surface energy of regions with spherical packing (the molecules are perpendicular to the surface) and confocal packing (the molecules are parallel to the surface). The second factor is the difference in the energy of elastic distortions of the two types of region, the energy density being determined by the values of the principal radii of curvature of the layers: $f \sim (1/R_1 + 1/R_2)^2$. For spherical layers $R_1 = R_2 > 0$, while for Dupin cyclides R_1 and R_2 have opposite signs ($R_1 R_2 < 0$), and as a consequence the elastic energy can be reduced. Thus, to determine ρ^* it is necessary to consider the total drop energy, consisting of a volume contribution $\mathcal{F}_V$ and a surface con-
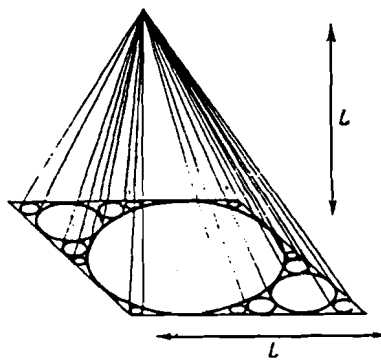

FIG. 2. Iterative filling of a region of space in the form of a pyramid by a family of contiguous confocal domains with a common apex at the apex of the pyramid.
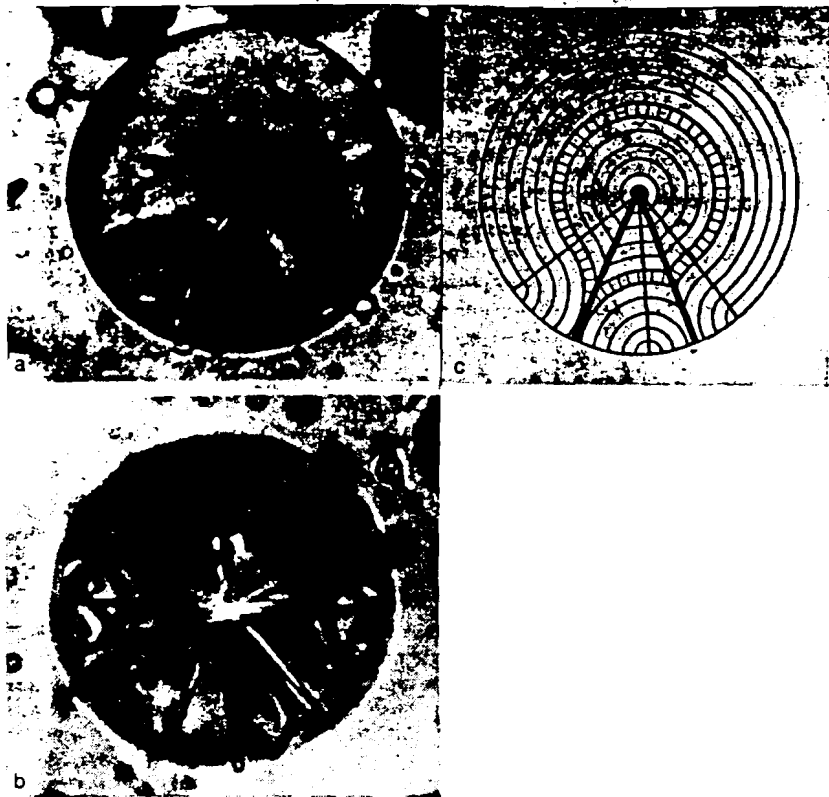
FIG. 3. Structure of spherical smectic-A drops suspended in the glycerine-lecithin matrix M_1: a,b) microphotographs of drops in transmitted polarized light (the analyzer is absent); c) scheme, corresponding to microphotograph (a), of the packing of the layers in the drop (section through the center of the drop).

tribution $\mathcal{F}_s$. To calculate $\mathcal{F}_V$ and $\mathcal{F}_s$ it is necessary to find a number of functions characterizing the filling of a spherical surface by an iterative system of contiguous circles of different radii ρ.

The initial stage of the interation consists in specifying a regular arrangement, on the sphere, of several equal contiguous circles of large radius. The problem is solved most simply for four, six, or twelve initial circles.[8] Then, in the gaps between the large circles, smaller ones are inscribed, and so on.

It is most important to know the total number $m(\rho)$ of circles occupying the sphere after each successive stage of the iterations. It is obvious that the number $m(\rho)$ is a function of the ratio R/ρ of the drop radius R to the radius ρ of the smallest circle. The determination of this function is a nontrivial problem not only for a spherical surface but even for a plane (the so-called Apollonius problem; see, e.g., Ref. 9). In the approximation $\rho \ll L$, where L is the characteristic size of the region being filled, it has been shown by computer-modeling methods that in planar geometry[4,9]

$$m(\rho) = A(L/\rho)^n, \qquad (2)$$

where A and n are numerical constants; the scaling index n takes the value $n \cong 1.3$ (for convenience of the calculations we take[4] $n = 4/3$). Since the character of the dependence is determined principally by the final stages of the iterative filling ($\rho \ll L$), we can regard formula (2) as being valid also for the filling of a spherical surface: For $\rho \ll R$ the geometry of a sphere differs little from the geometry of a plane. Therefore, we shall assume that the value $n = 4/3$ of the scaling index will also be applicable for a sphere.

This makes it possible to use, for the subsequent calculations, the results obtained for planar geometry in Ref. 4, in which it was shown that the elastic energy of the iterative system of confocal domains, written with allowance for the energy of the cores of the line defects is $\mathcal{F}_0 \cong K_{11}L^n\rho^{1-n}$, and the residual area not occupied by the bases of domains after the next iteration is $S_0 \cong L^n\rho^{2-n}$. Consequently, the volume energy $\mathcal{F}_V$ can be represented in the form of the sum of $\mathcal{F}_0$ and the energy associated with bending of the regions with spherical layers:

$$\mathcal{F}_V \approx K_{11}R^n\rho^{1-n} + 2K_{11}R^{n-1}\rho^{2-n},$$

and the surface energy can be represented as

$$\mathcal{F}_s \approx R^n\rho^{2-n}\sigma_\perp + (4\pi R^2 - R^n\rho^{2-n})\sigma_\parallel.$$

In writing this expression for $\mathcal{F}_s$ we have taken into account that on the boundaries of regions with spherical packing the surface tension is $\sigma_\perp$, and on the boundaries of confocal domains the surface tension is $\sigma_\parallel$ (we neglect small deviations of the molecular orientation from the tangential direction at the bases of the domains); in addition, L has been replaced by R, and K_{11} denotes the Frank bend modulus. Minimizing the sum $\mathcal{F}_V + \mathcal{F}_s$, we obtain the equilibrium value of ρ:

$$\rho^* = R(4 + 2R\Delta\sigma/K_{11})^{-1}. \qquad (3)$$

As can be seen from formula (3), the pattern of the space filling depends in an essential way on the magnitude of the anisotropy $\Delta\sigma = \sigma_\perp - \sigma_\parallel$ of the surface tension. Despite the fact that formula (3) was obtained with neglect of certain numerical coefficients, it correctly reflects the character

FIG. 4. Texture of a smectic-A liquid crystal (4-n-octyloxy-4-n-heptyl-α-cyanostilbene) in a planar sample with two glass surfaces.

of the dependence of ρ^* on the parameters $\Delta\sigma$, R, and K_{11}. If $\Delta\sigma \lesssim -K_{11}/R$ the iterative system does not arise and the drop has a structure of the type II, in which the molecules are oriented along the normal to the surface. For $\Delta\sigma \gtrsim -K_{11}/R$ the appearance in the drop of an iterative system of confocal domains, the smallest of which has a base of radius ρ^*, becomes energetically favored. As $\Delta\sigma$ increase, the critical radius gradually decreases from $\rho^*_{max} \sim R$ to $\rho^*_{min} \sim a$. The estimate $\rho^*_{min} \sim a$ follows from the fact that we have no grounds to assume that the energy of the interaction of the SALC molecules with the molecules of the matrix is appreciably greater than the energy of the intermolecular interaction in the SALC, and this implies that $\Delta\sigma$ also cannot be appreciably greater than K_{11}/a.

If the values of ρ^*, R, and K_{11} are known from experiment, the formula (3) can be used to estimate the quantity $\Delta\sigma$, which is an important characteristic of the surface of the SALC. For the investigated drops with the type-III structure in the glycerine-lecithin matrix M_1 (Fig. 3c), $\rho^* \cong 10$ μm, $R \cong 100$ μm, and $K_{11} \cong 10^{-11}$ N, and, consequently, $\Delta\sigma \cong 3 \cdot 10^{-7}$ N/m. In drops with the type-I structure, suspended in the pure glycerine matrix, it appears that the iterative process is truncated at much smaller values $\rho^* < 1$ μm, and, consequently, on the boundary of the SALC with the glycerine, $\Delta\sigma > 5 \cdot 10^{-6}$ N/m.

We now turn to the most frequently encountered experimental situation, in which the SALC has a planar shape, and show that the principal features of the space-filling pattern for drops are preserved in this case too.

3. TEXTURES OF PLANAR SAMPLES

The model for the filling of drops can be carried over most obviously to the geometry of an SALC sample enclosed between two cover glasses and containing three air bubbles (Fig. 4). It can be seen that the confocal domains are combined into three families around each of the bubbles. Here the domains of each of the families are oriented in such a way that the continuations of all the hyperbolas converge to the same point—namely, the center of the corresponding air

bubble. Thus, the given structure is a part of the structure of the type-III drops considered above.

We shall consider the character of the space filling in the most frequently encountered SALC texture—namely, a polygonal texture (Fig. 5a). This figure shows a texture of the liquid crystal CF_3-C_6H_4-$CH = N$-C_6H_4-C_4H_9 (fluorinated MBBA), which forms a smectic-A phase in the range 43–49 °C. The substance was placed between cover glasses, onto which a thin layer of the glycerine-lecithin matrix M_1 had first been deposited. The layer thickness was $L = 200$ μm. The use of this way of preparing the cover glasses made it possible to obtain, in a reproducible manner, qualitative polygonal textures in which most of the confocal domains had a toroidal configuration (the latter is evidently connected with the creation of conditions for the degenerate orientation of the SALC molecule at the surface, and with the lowering of the azimuthal part of the energy of cohesion). We note that for planar surfaces prepared in a different way, the polygonal textures preserve their main features; only the quantity ρ^* is subject to changes. The sizes of the polygons are close to the sample thickness. Their edges are slightly curved and are formed by segments of lines of confocal pairs. The families of polygons in the texture are localized both on the upper and on the lower surface of the sample.

Each polygon is the base of a pyramid whose apex lies on the opposite bounding surface. Here the apices of the pyramids whose bases lie in the lower plane coincide with the apices of polygons lying in the upper plane, and vice versa.

A general scheme for the filling of a planar sample by such pyramids was proposed by Bragg.[3] If for simplicity we choose pyramids with square bases, then the whole space can be divided into two sets of pyramids (of the type $H(ABCD)$ and $A(EFGH)$ in Fig. 6a), with apices on opposite surfaces of the sample, and also into a complementary family of tetrahedra of the types $ABGH$ and $ADEH$ in Fig. 6a, which fill the regions between the pyramids. The upper and lower edges of each tetrahedron are simultaneously edges of polygons in the bases of the pyramids. They can be regarded as parts of lines of a confocal pair; the layers within each tetrahedron have the shape of Dupin cyclides.[3,4] By definition, the straight lines joining any points of a confocal pair are normal to the Dupin cyclides. Consequently, each tetrahedron is a part of a confocal domain (Fig. 6b), and the smectic-A layers intersect its faces at a right angle (for more detail, see Ref. 3).[1]

From the above account it follows that the problem of the space filling in a polygonal texture reduces to the problem of the filling of pyramids. As can be seen from Fig. 5a, the filling of pyramids takes place analogously to the filling of drops. In the polygons (the bases of the pyramids) are inscribed the ellipses of an iterative family of confocal domains: one or more at the center, and smaller ones in the corners of the polygons and in the gaps between the large domains, and so on. The hyperbolas of all domains converge at the apex of a pyramid. As in the situations considered above, the iterative filling is truncated at domains with a certain minimum size ρ^*. The remaining gaps inside the pyramid are occupied by spherical layers with a common center of curvature at the apex of the pyramid. The latter statement
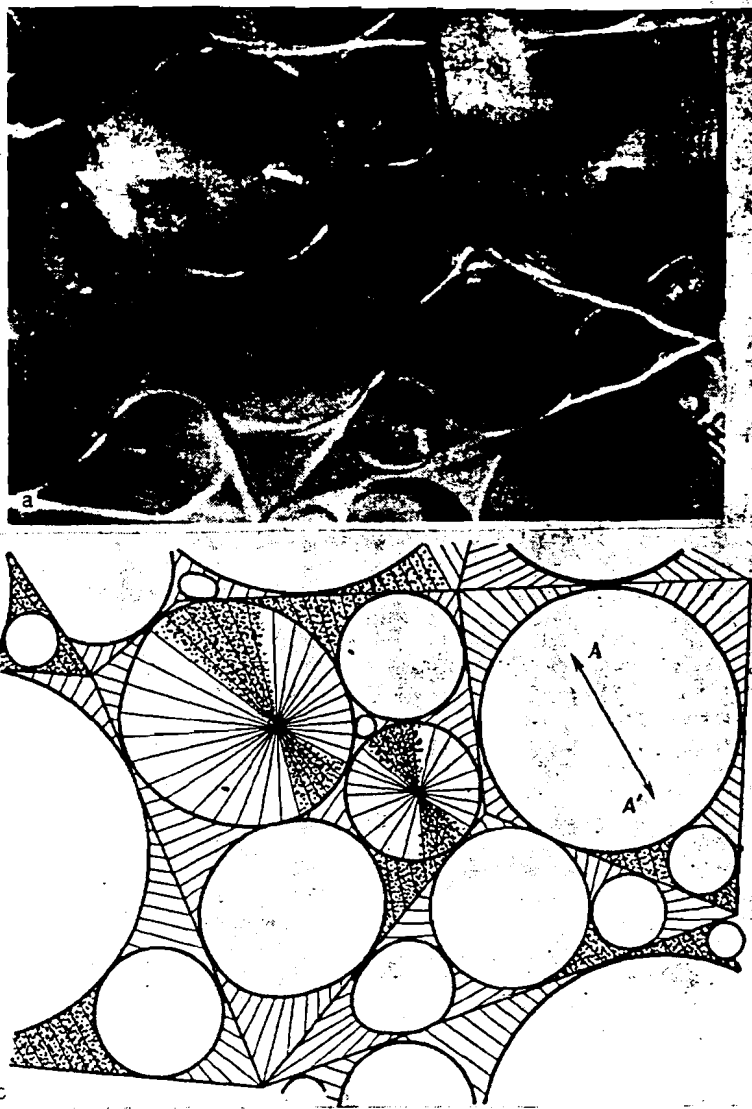
FIG. 5. Polygonal smectic texture in a sample of thickness 200 μm: a) microphotograph of the upper surface of the texture, in transmitted polarized light with one analyzer; b) the corresponding scheme of the packing of the bases of the confocal domains into polygons—the bases of the pyramids. The distribution of the optical axes on the surface is indicated by the thin straight lines, and the regions of darkening, in which the optical axis is parallel to the plane of polarization of the analyzer, are shaded by dots.

can be checked by studying the pattern of the extinction in a polygonal texture in observation in a microscope with one analyzer. It is known that the regions of extinction should be localized in those parts of the texture where the optical axis (the normal n to the layers) is parallel to the plane of polarization of the analyzer (see, e.g., Ref. 3). The distribution of the optical axes for a system of layers in the plane of the polygons in Fig. 5a is presented in Fig. 5b. The optical axes in each confocal domain in this plane converge at one of the poles of the ellipse, and the projections of the optical axes of the free parts converge at the point of the projection of the apex of the pyramid on the plane, if these parts are occupied by spherical layers. This arrangement of the optical axes is indeed observed experimentally. From a comparison of Fig. 5a and Fig. 5b it can be seen that the character of the space filling on the microphotograph corresponds to the scheme of filling by spherical layers. The individual deviations in the degree of darkening in the upper part of the microphotograph are due to light ovals caused, apparently, by variations of the glycerine layer. The critical scale of the iterative space filling is determined analogously to the expression (3), if R

is replaced by the characteristic size L of a polygon.

It remains to convince ourselves that the smectic layers can cross smoothly from pyramid to pyramid through regions of tetrahedra, without creating defect walls on the way. This is in fact the case. First, in the above-described filling of a pyramid, the side faces of the pyramid are intersected by a family of spherical layers at a right angle; the layers of a family of Dupin cyclides, belonging to the neighboring tetrahedron, also approach the faces of the pyramid at a right angle. Secondly, in the region of the crossing, e.g., in the plane ABH (Fig. 6d) the layers of both families have a common center of curvature at the apex of the pyramid (the point H for the plane ABH in Fig. 6d).

The above examples of the filling of spherical drops and polygonal textures permit us to conclude that in other experimental geometries the general features of the proposed model will be preserved. In particular, in the fan textures that are formed in thin samples ($L \sim 10\mu$m), it is possible to observe how the hyperbolas lying in the plane of the sample converge at the same point—the center of the spherical domain whose layers fill the gaps between the confocal do-
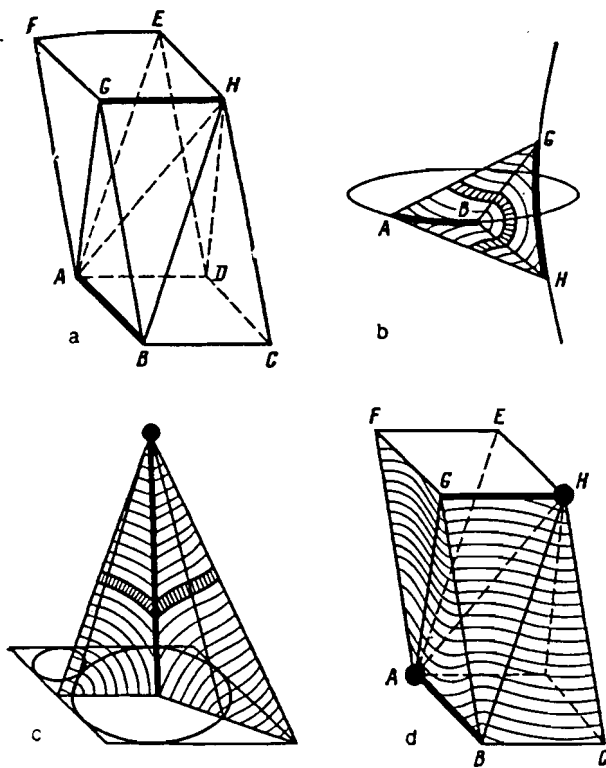
FIG. 6. General scheme of space filling by smectic layers: a) division of a parallelepiped (which can be used to fill space by means of parallel translations) into a pair of pyramids $H(ABCD)$ and $A(EFGH)$ and a pair of tetrahedra $ABGH$ and $ADEH$; b) filling of a tetrahedron $ABGH$ by a single family of Dupin cyclides intersecting the faces of the tetrahedron at a right angle; c) filling of a pyramid by a single family of spherical layers, into which confocal domains are built smoothly; d) scheme of a smooth crossing of layers from a pyramid to a tetrahedron within one parallelepiped.

mains. The only exceptions, evidently, are textures of parabolic domains, in which the space filling can be produced by a regular lattice of domains of the same size.[11]

4. CONCLUSION

For the examples of a number of SALC textures we have considered the pattern of the space filling by flexible layers of constant thickness when this pattern satisfies conditions on the orientation of the layers at the surface. The principal features of the pattern reduce to the following.

On large scales, comparable with the characteristic size of the system, the space filling is realized by an iterative system of contiguous confocal domains of successively decreasing sizes. The size of the base of the smallest domains is determined principally by the conditions on the orientation of the layers at the surface of the system. On smaller scales the hierarchy of confocal domains is replaced by a new type of packing; all the remaining free gaps are filled by spherical layers.

As follows from the above account, in space filling by smectic layers the only defects are point radial "hedgehogs" and pairs of lines in the form of an ellipse and a hyperbola (the latter, strictly speaking, are not topologically stable singularities). At the same time, in real experimental conditions the structure of the confocal and spherical domains can

be distorted, first because of violation of the conditions (1) requiring equal spacing of the layers (this corresponds to the appearance of edge and screw dislocations[10,12]), and secondly because the Dupin cyclides tend to adopt the shape of minimal surfaces ($R_1 = -R_2$), as has probably been observed in certain cases for lyotropic[13] and cholesteric thermotropic[14] phases. However, these distortions do not introduce great changes into the large-scale pattern of the space filling.

The problem of space filling by flexible layers has a number of analogies, noted previously in Refs. 4 and 5. In connection with the distinguishing feature of the proposed model (the presence of a characteristic critical scale ρ^* substantially exceeding, in the general case, the molecular sizes), it is of interest to give a further brief discussion of some of these analogies.

The situation closest to the model under consideration is the situation with "branching" of domains of the normal phase in the intermediate state of superconductors, in which, as they approach the boundary of the sample, the domains experience a series of successive branchings into domains of smaller sizes.[15] However, this process, like the iterative space filling in a smectic, is not infinite and ceases at scales much larger than the coherence length. The cessation of the branching is caused by the decrease in the magnitude of the critical field when the domain sizes decrease.[16] A comparison that is becoming better founded is that between the packing of smectic layers in space and the pattern of turbulent flow, in which pulsations of different sizes, from the largest, of the order of the characteristic size of the system, to the smallest, determined by the so-called internal scale of the turbulence,[17] are present.

Finally, a nontrivial analogy is that between the filling of space by confocal domains of a spherical drop and the faceted structure of the eyes of insects.[5] The role of the confocal domains in the latter case is played by the conical formations (ommatidia), which emerge onto the almost spherical surface along the sides of the insect's head.[18] For a number of physical reasons, described in detail by Feynman *et al.* in Ref. 18 and associated principally with features of the refraction of light, there exists a critical ommatidium radius that ensures optimal sharpness of vision. It is interesting that this size (30–35 μm) is close to the characteristic radius in the drops with structure of type III described in Sec. 2. It seems logical to assume that the simplest way for forming a faceted structure is to create conditions similar to those described for drops in Sec. 2.

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[1] Opposite sides of polygons can also be two hyperbolas; such a pair of sides is not confocal, and in the filling of the corresponding tetrahedron a system of screw dislocations arises.[10]

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