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Focal conic faceting in smectic-A liquid crystals

J B Fournier and G Durand

Laboratoire de Physique des solides, bât 510, 91405 Orsay Cedex, France

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Abstract. — Smectic-A nucleating in the isotropic phase presents various bulk focal conic defects close to the interface, within the « bâtonnets » first described by Friedel and Grandjean in 1910. We experimentally study the stability of the « bâtonnets » in a high-frequency aligning electrical field. Although the external shape appears out-of-equilibrium, the focal conic texture is found stable. We build a model explaining the general interfacial focal conic texture. It participates in the minimization of the interfacial energy by orienting the smectic layers almost perpendicular to the interface, above a critical size \( l_0 \) comparing layer curvature and surface energies. This model explains new arborescent textures that we observe inside capillary tubes. We then discuss equilibrium shapes of size \( L \). We discuss three regimes \( L < l_0 \) yields 3D-cristal-like « Wulfs » shapes. For \( L > l_0 \), equilibrium shapes must contain focal conics, although we cannot solve the general solution. In the asymptotic limit \( L \rightarrow \infty \), we predict fractal textures of dimension \( 1 < d < 2 \) inside « optimal » configurations, spherical as for 3D-liquids. Finally, focal conics can be considered as « bulk facets » which pile up to decorate « bâtonnets » and more general interfacial textures. Focal conics appear then as intrinsic defects of Smectic-A materials in contact with their isotropic phase.

1. Introduction.

Smectic-A (Sm-A) are lamellar liquid crystalline phases [1]. In the ground state, an « ideal » Sm-A phase consists in a uniform piling of planar, parallel and equidistant layers of molecular thickness. In practice, Sm-A phases usually present bulk distorted textures in which the layers, although remaining equidistant, can be violently curved. These macroscopic defects, called « focal conics » were first studied by Friedel and Grandjean [2]. They consist in delimited bulk domains inside which the Sm-A layers are curved in the vicinity of two disclination lines, an ellipse and the conjugated « confocal » hyperbola.

Usually, focal conics are produced by external constraints forcing layer curvature. In most cases, the curvature is induced to fulfill antagonistic orientational boundary conditions. These antagonistic « anchorings » generally occur on solid extrinsic substrates [2], or between smectic drops and amorphous liquid matrixes, on mixture interfaces [3]. However, it has recently been shown that the surface energy anisotropy of interfaces intrinsic to the liquid crystalline material could also induce focal conic defects [4] in a particular planar geometry, in which the Sm-A phase is sandwiched between air and the isotropic phase of the material, a first-order textural instability creates free floating focal conic networks. The focal conic curvature is produced by the antagonistic surface anisotropies of the two interfaces, the Sm-
A/air interface favors parallel layer/surface orientation and the intrinsic Sm-A/isotropic interface favors perpendicular layer/surface orientation.

More generally, it is interesting to discuss whether a similar instability would occur, in the absence of any air phase, on intrinsic Sm-A/isotropic interfaces of various shapes, i.e. to study the problem of the equilibrium shape of a Sm-A volume nucleated inside the isotropic phase. Indeed, it is known since Friedel and Grandjean’s studies in 1910, that the nucleation of Sm-A inside Isotropic phase gives rise to various elongated shapes, the «Bâtonnets» [2], decorated by more or less complicated focal conic textures. However, whether the «origin» of these focal conics is statical or dynamical, or whether they are stable or metastable has never been established.

In this paper, resuming the anisotropy of the Sm-A/isotropic interfacial energy as the leading mechanism, we study the equilibrium shapes of Sm-A nucleated inside the Isotropic phase of the liquid crystalline material. After recalling in part 2 the focal conic structure and the associated energy, we establish in part 3, the «Wulf-like» elongated shapes that would correspond to ideal strictly undistorted Sm-A liquid crystals nucleated inside the isotropic phase. Then, we discuss the associated elastically relaxed configuration and show that the amplitude of the relaxation should be infinitesimal. Generalizing the model of reference [4] for Sm-A plates, we construct a configuration containing a single focal conic defect which is significantly less energetical, above a typical size, than the previous Wulf solution. Equilibrium shapes must then contain focal conics. In part 4, we experimentally study the stability of the «bâtonnets» focal conics. We show that although the external shape is probably out of equilibrium, according to a «1D-crystal behavior», the bulk focal conic can be considered as an equilibrium texture. In consequence, in part 5 we consider interfaces of arbitrary shapes. We establish, in the case of large interfaces, a general procedure to build focal conic textures relaxing the anisotropic interfacial energy. We then discuss the case of the «bâtonnets». In part 6, we study asymptotic equilibrium shapes in the limit of large Sm-A volumes and show that it must correspond to spheres with a radial «Apollonius» [5] network of focal conics. Finally, we show in conclusion that focal conics can be geometrically considered as the «volumic facets» of Sm-A equilibrium shapes, by analogy with the surface facets of usual 3D-crystals. A partial summary of this work was recently presented in a workshop [6].

2. Focal conic bulk defects.

An «ideal» Sm-A phase, without any defects, consists in a one-dimensional (1D) piling of 2D liquid layers, at the same time planar, parallel and equidistant (Fig 1) Inside each layer of molecular thickness a (a ~ 30 Å for usual thermotropic Sm-A), rod-like molecules are on the average oriented in the direction of the normal n to the layers (nematic ordering of director n, n² = 1). The specific properties of smectic phases are mainly due to the fact that they are low-dimensional crystals (1D-crystals and 2D-liquids). For instance, unlike 3D-crystals which always consist in domains of uniform orientation separated by grain boundaries, real Sm-A phases are never quite ideal but do usually show macroscopic bulk defects, the so-called focal conics, in which the Sm-A layers are no longer planar but violently curved. This property is characteristic of low-dimensional crystals, which can exhibit large bulk distortions of pure curvature strictly leaving the 1D-crystal undilated. In the case of 3D-crystals, however, all types of bulk distortions dilate the 3D-lattice.

The free energy density \( f_d \) associated with the bulk elastic distortions of Sm-A phases contains two terms [1]:

\[
f_d(r) = \frac{1}{2} K \{c_1(r) + c_2(r)\}^2 + \frac{1}{2} B \epsilon(r)^2
\]

(1)
The first term describes the curvature energy (\(c_1\) and \(c_2\) being the local principal curvatures of the Sm-A layers, \(K \sim 10^{-6}\) cgs); the second term describes the energy associated with layer dilation (\(\varepsilon\) stands for the relative layer thickness variation, \(B \sim 10^8\) cgs). Note that \(B \sim K/\lambda^2\) defines a «penetration» length \(\lambda\) [1], which compares with a few molecular lengths far from second-order phase transitions.

With simple dimensional arguments, let us first recall why pure curvature defects do actually occur in Sm-A. We consider a one length problem in which the Sm-A layers are curved with a typical curvature radius \(R \sim L\) inside a volume \(\sim L^3\) (Fig. 2a) The total integrated corresponding energy writes \((K/L^2) L^3 \sim KL\) and grows linearly with the size \(\sim L\) of the domain. Note that if \(R\) goes to zero inside the volume, a Log \(L\) factor appears because of the curvature divergence. However, such a modest factor will not change the discussion qualitatively, as shown later on. This, indeed, is a remarkable point that curvature volumic distortions behave as «lines». Therefore «focal conic» pure curvature defects, of

Fig. 1 — Schematic representation of an «ideal» Sm-A phase

Fig. 2.— Elastic distortions in a one length \(\sim L\) problem. a) The Sm-A layers are curved \(\sim 1/L\) inside a volume \(\sim L^3\). b) They are also generally dilated if no precaution is taken
energy roughly $\sim KL$, are less energetical than grain boundaries, whose energy obviously scales $\sim L^2$. In the case of 3D-crystals, grain boundaries are created to separate domains having different orientations. With our previous argument, smectics will prefer curvature to grain boundaries to connect such different domains. In practice, it is often the case that an antagonistic anchoring of the layers on the boundary plates induces a macroscopic layer curvature $\sim 1/L$ on a length $\sim L$. Now, a difficulty arises since curving arbitrarily the layers will also arbitrarily dilate them (Fig. 2b). Let us then compare curvature distortions with dilative distortions in the same one length problem (Be$^2$) $L^3 \sim KL$ gives $\varepsilon \sim \lambda /L$, which corresponds to the maximum layer thickness variation in the volume $\sim L^3$, if we require that dilation energy should not exceed curvature energy. Since $\lambda$ is molecular, $\varepsilon$ must vanish for macroscopic curvature; this of course comes from the fact that dilation energy scales $\sim L^3$ and pure curvature only scales $\sim L$. Since macroscopic curvature $\sim L$ cannot «suffer» a dilation greater than $\varepsilon \sim \lambda /L$ (e.g. $\varepsilon \sim 10^{-3}$ for $L \sim 10 \text{\mu m}$, far from second-order phase transitions), it is a very good approximation to use the geometrical description, i.e. to assume strictly $\varepsilon = 0$, excluding layer dilation [7].

We now recall the focal conic structure [8], using our previous arguments which general structure can we build with families of curved but equidistant layers? If one starts with any general (undulated) surface and try to build parallel and strictly equidistant surfaces, the process will rapidly stop because singularities, where the layers are no more defined, will appear on surfaces [8] (Fig. 3). This is not acceptable because the energy $\sim L^2$ of such surface singularities compares with that of grain boundaries. To build structures of linear energy $\sim L$, the rules must be revisited in the following way: which general structure can we build if we curve the Sm-A layers with the constraints of keeping them strictly equidistant and creating no more than point or line singularities? For point singularity, the solution is trivial and consists in folding the layers on concentric spheres, the « disclination» point singularity being the common center of the spheres (Fig 4a). In the case of line singularities, it can be shown [8] that, in the most general case, they must be an ellipse ($E$) and the conjugated « confocal » hyperbola ($H$) in the perpendicular plane (Fig. 4b) The term « confocal » refers to the property that the focus of one conic coincides with the summit of the other and vice

![Figure 3](image-url)
versa. These confocal conic singularities are Volterra disclination lines of the (nematically oriented) smectic ordering. The ordering (isotropic or nematic) in the core of these lines is not known. At least the smectic ordering does melt.

The bulk Sm-A layers are built in such a way that their local normal \( \mathbf{n} \), which is also the nematic director, lies on the fictitious lines joining any point \( \mathbf{M} \) on the ellipse to any point \( \mathbf{M}' \) on the hyperbola (Fig 4b). The simplest focal conic has a revolution symmetry and corresponds to the degenerated case in which the ellipse becomes a circle and the hyperbola becomes the perpendicular conjugated straight line (the revolution axis) passing through the center of the circle. The Sm-A layers are then concentric tori folded around the circle and intercepting the straight line with an angular discontinuity when the radius of the tori becomes larger than the circle radius. The general ellipse-hyperbola focal conic structure can be easily derived by continuous deformation from the previous case, i.e. the Sm-A layers take the general shape of the so-called Dupin's cyclides corresponding to tori-like surfaces whose «radii» increase in the direction of the ellipse focus not occupied by the hyperbola.

A difficult problem, already discussed by many authors [2, 3, 5, 8, 9], concerns the filling of space with focal conic defects. The principal problem is to avoid strains between neighbouring focal conics. The main results can be summarized as follows: one can define conical domains as the focal conic texture limited inside a cone whose apex \( \mathbf{O} \) lies on the hyperbola and whose base is the ellipse (Fig. 5a). It is a strong geometrical property of «confocal» conics that such cones are always cones of revolution. Since the molecules are parallel to the cone generatrices everywhere on the cone, no disclination strains arise if the cones are associated tangentially along their generatrices [8] (Fig 5b). Such cones define a radial distribution around their common apex \( \mathbf{O} \) and the interstices between them can be filled, without strains, with spherical layers [9] also centered on \( \mathbf{O} \). The further filling associations depend on the particular boundary conditions and boundary shapes.

Let us come back to the energetics of focal conics. Since the curvature diverges near the disclination lines, it is now no longer obvious that a focal conic domain of size \( \sim L \) (size of the ellipse) corresponds to an energy \( \sim KL \). The length \( \xi \) (the smectic coherence length) of the cores around disclination lines appear as a natural cut-off for the curvature.
energy integration. Although the problem is no longer a one-length problem, it has been shown [10] that the core size $\xi$ only appears in a slowly varying corrective factor $\sim \ln (L/\xi)$. The focal conic energy is the sum of three terms: the energy $W_b$ of the integrated bulk curvature and the energies $W_e$ and $W_h$ of the respective ellipse and hyperbola cores of disclination. In the case of a degenerated focal conic of revolution, the bulk term $W_b$, integrated in a cylindrical domain of revolution whose base is the circle of radius $r$, is written [10] $W_b \sim K r \ln (r/\xi)$. Traditionally [1], the core disclination of the ellipse, which is topologically melted everywhere, gives a contribution of order $\sim K$ per unit length (curving an angle unity over a distance $\sim \xi$ must compare with melting). This gives $W_e \sim K \ell_e$, in which $\ell_e$ represents the ellipse length. However, a more precise description should take into account, for various eccentricities, the dependence of the proportionality coefficient along the line. The case of the hyperbola disclination line is more delicate, it has been shown [11] that both the core disclination and its energy should vanish far from the ellipse since the angular discontinuity becomes infinitesimal and can be elastically relaxed. It is therefore a usual approximation [5] to count only the significative part of the hyperbola, whose length compares with that of the ellipse. The energies of the two disclination lines are then roughly proportional. The total energy, integrated inside an oblique cylindrical domain having the hyperbola direction and the ellipse base, can then be written as

$$W \sim K \ell_e \{\alpha + \beta \ln (\ell_e/\xi)\},$$

the proportionality coefficients $\alpha$ and $\beta$ depending on the eccentricity [10]. Formula (3) should be regarded as valid in the limit of very long hyperbolae. If the hyperbola length compares with the ellipse length, it can be convenient [4] to treat them as independent variables, i.e. to add a term $\sim K \ell_h$ proportional to the hyperbola length. The previous general results on focal conics will be useful to discuss the part played by bulk distortions in the equilibrium shapes of Sm-A nucleated inside its isotropic phase.

3. Equilibrium shapes.

Surface energy anisotropies play an important role in determining equilibrium shapes. Observing Sm-A phases, Friedel first guessed [12], from the shape of the smectic germs
(« bâtonnets »), the tendency of the layers to orient normal to the Isotropic/Smectic-A interface. He also noticed the tendency of smectic layers to align parallel to air drop surfaces (« gouttes à gradians ») He was however puzzled, in the first case, that a surface energy minimum could exist in a non-crystalline direction. Later on, X-ray experiments [13] demonstrated the tendency of the Sm-A layers to align parallel to air interfaces More recently, quantitative temperature measurements of the Sm-A/air surface energy were performed on equilibrium facets for free drops laying on a solid substrate [14]. The Sm-A/air parallel alignment corresponds to the expected crystalline behavior. Conversely, as was quantitatively shown recently by the textural instability of reference [4], interfaces with the isotropic phases of Sm-A materials do present the opposite tendency: the smallest surface energy corresponds to the perpendicular layer/interface orientation. The experiment of reference [4] studied the behavior of a Sm-A plate sandwiched between two parallel interfaces Sm-A/air on one side and Sm-A/Isotropic on the other. Above a thickness threshold of the Sm-A plate, a textural instability occurs, creating free floating hexagonal networks of focal conics, whose bulk curvature is induced to fulfil the two antagonistic interfacial anisotropies.

In the absence of any air interface, a frustration persists [15]: a closed Sm-A volume, completely immersed inside the isotropic phase, cannot have both the preferred ideal bulk ground state (parallel planar equidistant layers) and the preferred perpendicular layers/interface orientation everywhere along the boundary. It is then of some interest to examine which compromise would be chosen in this frustrated situation. In particular, we wonder whether intrinsic focal conics should be present or not at equilibrium: what would be the equilibrium « shape » of a Sm-A closed volume having nucleated inside the isotropic phase?

We first recall some well-known simple results on crystal equilibrium shapes. The anisotropic surface energy \( \gamma(\theta, \phi) \) of crystals, function of the relative bulk/surface orientation usually defined by the zenithal and azimuthal angles \( \theta \) and \( \phi \), leads to equilibrium shapes with facets and, more generally, elongations, which differ strongly from the spherical shapes corresponding to \( \gamma \) isotropic. In the simplest model, to determine the equilibrium shapes, one must minimize the surface energy for a given fixed total volume, with respect to the surface shape, i.e. solve Wulff's problem:

\[
\int \gamma \, dS + \alpha \, V = \text{min}.
\]

The corresponding variational formulation leads to the well-known Wulff's construction [16]. In such a description, for 3D-crystals, the bulk is strictly assumed to be « ideal », i.e. the eventuality of bulk elastic distortions, which involve too high energies, is not considered. Therefore, since \( \gamma \) is the only physical quantity in the problem, no characteristic length appears and Wulff's solutions are homothetic (the solution for a given volume \( V \) is deduced from the one corresponding to a volume \( V \) by a simple homothetie, i.e. has the same « shape »). Usually, in the case of 3D-crystals, the function \( \gamma(\theta, \phi) \) presents derivative discontinuities near the orientations close to crystalline planes. These discontinuities are responsible for the existence of facets in equilibrium shapes. However, real crystal facets do not correspond to equilibrium but are rather due to dynamic effects [17], except in very few cases as in solid helium [18].

To discuss the case of Sm-A, let us first establish the corresponding Wulff's solution. In agreement with reference [4], we assume \( \gamma \) to be anisotropic, the minimal value \( \gamma_\perp \) corresponding to layers perpendicular to the Sm-A/Isotropic interface, and the maximal value \( \gamma_\parallel \) corresponding to layers parallel to it. In addition (and also by lack of information), we assume \( \gamma(\theta) \) to be a smooth function of the angle \( \theta \) between the normal to the Sm-A layers...
and the normal to the interface. From uniaxial symmetry, \( \gamma \) is independent of \( \varphi \). Wulf's solution must then be a defined homothetical shape of revolution, the normal to the monocrystalline layers' piling being parallel to the axis of elongation in order to increase the lengths of the low energetical surface parts (Fig. 6a) Let us call \( L \) the length of the long axis, i.e. the scaling length. Due to the homothetical property, the total surface of the solution is \( S = \alpha L^2 \), where \( \alpha[\gamma] \) is a pure constant (\( \alpha \sim 1 \)). The total energy \( W_w \) of Wulf's solution contains only a surface term. Integrating the interfacial energy yields

\[
W_w = \alpha \bar{\gamma} L^2
\]  

(4)

where \( \bar{\gamma} \) is a constant angular mean value of \( \gamma \). Wulf's solution explores the function \( \gamma(\theta) \) by locally changing the surface orientations with respect to the fixed bulk axis. In fact, the interfacial energy is a function of the surface/volume respective orientation which can also be modified by locally changing the bulk axis directions with respect to the surface, i.e. introducing bulk distortions. In the case of 3D-crystals which are very « stiff », it is a very good approximation to neglect these degrees of freedom. However, Smectics which are only 1D-crystals (and 2D-liquids) are much « softer » and can exhibit a very different behavior.

![Diagram](image)

Fig 6 — a) Wulf's solution corresponding to an assumed « ideal » strictly undistorted Sm-A bulk b) Previous configuration elastically relaxed \( \sim \delta \theta \) As soon as \( L \) is macroscopic, \( \delta \theta \) is found infinitesimal

Solving the general problem including bulk distortions, i.e.

\[
\int \gamma \, dS + \int f_d \, dV + \alpha V = \min
\]  

(5)

is a difficult problem, the solutions of which are unknown. In principle, \( f_d \) must describe all the possible discrepancies with respect to the ideal bulk equilibrium ground state [1], i.e.

\[
f_d(r) = f_{el}(r) + \delta f \{ \psi(r) - \psi_{eq} \}
\]  

(6)

The first term is the elastic free energy density associated with curvature and dilation of the
Sm-A layers (see Part 2) and the second one describes an eventual melting of the Sm-A order (e.g. near disclination lines since we are not concerned with dislocations which are macroscopic defects).

Although the solutions of equation (5) are unknown, it is possible to make a semi-quantitative discussion. Let us see how Wulff's solution, which strictly assumed rigid layers, now adapts if we reintroduce the finite Sm-A elasticity. As the smectic layers are not everywhere perpendicular to the interface, a surface torque bends the layers symmetrically and drives the system towards an elastically relaxed configuration (Fig. 6b). Let us call $R = \mu L$ ($\mu < 1$) the revolution radius of Wulff's solution, $\theta_0$ a «mean» angle at the surface and $\delta \theta$ the «mean» bending angle. An elastic relaxation $\sim \delta \theta$ creates both curvature of «mean» radius $\rho \sim R/\delta \theta$ and dilation of «mean» amplitude $\varepsilon \sim R(\delta \theta)/L$. The corresponding energies, integrated in the volume $V = \beta [\gamma] L^3$, with $\beta \sim 1$, scale then as

$$W_K \sim \frac{1}{2} K \left( \frac{\delta \theta}{R} \right)^2 \beta L^3 \sim KL (\delta \theta)^2$$

$$W_B \sim \frac{1}{2} B \left( \frac{R\delta \theta}{L} \right)^2 \beta L^3 \sim BL^3 (\delta \theta)^2$$

As $L$ is always much larger than $\lambda$, the leading term is $W_B$ ($W_K \ll W_B$). Assuming $\gamma$ to have roughly the form $\gamma(\theta) \sim \gamma_\perp + \Delta \gamma \cos^2 \theta$, the energy of Wulff's solution is lowered by an amount

$$\Delta W \sim \alpha L^2 \Delta \gamma \{ \cos^2 (\theta_0 + \delta \theta) - \cos^2 \theta_0 \} + \frac{2\beta}{\mu^2} BL^3 (\delta \theta)^2$$

Minimization of equation (9) with respect to $\delta \theta$ yields:

$$\delta \theta \sim \frac{1}{4} \frac{\alpha \mu^2}{\beta} \sin 2 \theta_0 \left( \frac{\lambda^2}{\ell_0 L} \right) \sim \frac{\lambda^2}{\ell_0 L}$$

in which we have introduced both $\lambda \sim (K/B)^{1/2}$ and $\ell_0 \sim K/\Delta \gamma$. The constant $\lambda$ is known to compare to a few molecular lengths far from second-order phase transitions and the order of magnitude of $\ell_0$ is known as a result of the instability studied in reference [4], i.e. $\ell_0 \sim 0.1-1 \mu m$ for usual Sm-A phases, with $K \sim 10^{-6}$ cgs and $\Delta \gamma \sim 0.1$ cgs. It follows that above microscopic molecular sizes, $\delta \theta$ is infinitesimal and this relaxation irrelevant as in the case of 3D-crystals. This is due to the fact that this symmetric solution which can be obtained continuously from Wulff's one must involve dilation of the 1D-crystal. Dilation energy which scales $\sim L^3$ must be of infinitesimal amplitude to compare with interfacial energies scaling $\sim L^2$. To find configurations definitely less energetical than Wulff's solution, one must introduce only non-dilative bulk distortions. This is indeed possible, as we show now, in the case of low-dimensional crystals like smectics and corresponds to the pure curvature of focal conic defects.

We shall now exhibit a particular configuration with focal conic bulk distortions of energy $W_{FC}$ lower than the energy $W_B$ of Wulff's solution. We choose a particular configuration which has the same homothetic external surface than Wulff's solution and, in addition, we place a revolution focal conic (circle on the surface and conjugated straight line on the revolution axis) inside the bulk, according to figure 7a. Note that the symmetric up-side down focal conic is not introduced as will be discussed in part 5. $W_{FC}$ contains a surface term $W_s$ and a volume term $W_b$. As the external shape is chosen homothetically identical to Wulff's one, the surface energy is $W_s = \alpha \gamma_{FC} L^2$. $W_s$ is now significantly lower than Wulff's surface energy since the layer orientation fits better the extremity near the circle and is quite unchanged everywhere.
Fig 7 — Model configurations based on Wulff's solution with one revolution focal conic included. a) Perspective view of the disclination lines b) Cross section through the revolution axis. The Sm-A layers are now quasi perpendicular to the Sm-A/Isotropic interface near the circle E. H should be tilted to allow for another focal conic to relax the other end.

else (Fig 7b), i.e. $\gamma_{FC} < \tilde{\gamma}$ strictly. Therefore, since the focal conic energy $W_0$ is negligible compared to $L^2$ for $L$ large enough, i.e. $W_0 \sim \varepsilon(L^2)$, there exists a characteristic length $\ell_0$ above which the solution including the focal conic is energetically lower. To determine $\ell_0$, let us introduce explicitly the focal conic energy $W_0$ of order $(\text{Cte}) KR \ln (R/\xi) + (\text{Cte}) KR$, as previously discussed in part 2. The total energy can then be written as

$$W_{FC} \sim \alpha \gamma_{FC} L^2 + \{ \nu_1 + \nu_2 \ln (L/\xi) \} KL$$

(11)

where $\nu_1$ and $\nu_2 \sim 1$ are dimensionless numbers. Therefore, $L > \ell_0$ yields $W_{FC} < W_w$, $\ell_0$ being given by

$$\ell_0 \sim \frac{\nu_3}{\alpha \tilde{\gamma} - \gamma_{FC}}$$

(12)

where $\nu_3 = \nu_1 + \nu_2 \ln (\ell_0/\xi) \sim 1$. And again, we find back the typical size $\ell_0 \sim K/\Delta \gamma$, of order 0.1-1 μm, comparing the anisotropy of the surface energy with the Sm-A layer curvature constant. Note that in other contexts with fixed boundary shapes, such a characteristic length due to the competition between surface and bulk energies was first mentioned in reference [9] and then developed in reference [3]. The main difference here is that we discuss the intrinsic case of nucleation, in which both surface and bulk texture are free parameters.

We have therefore found a particular configuration containing one focal conic, which is energetically lower than Wulff's solution. This indeed is enough to ensure that equilibrium shapes must contain focal conics above the typical size $\ell_0$. However, the general equilibrium shape solution is probably much different from this simple one. In fact, because of $\ell_0$, the homothetical property disappears, and one should expect, for each volume $V$, one particular solution characterized both by its external surface shape and its bulk focal conic texture. One should then also expect first-order transitions between different classes of
solutions involving different focal conic arrangements, and all corresponding hysteresis phenomena. For instance, in the vicinity of $l_0$, an instability similar to that of reference [4] should occur to create the first focal conic from Wulff's solution. Unfortunately, the value $l_0 \sim 0.1-1 \mu m$ prevents from observing it directly under optical microscopy. Note that in the geometry of reference [4], it is because of the presence of the air interface that the instability threshold is raised up to a macroscopic value. However, as known since 1910, focal conics are indeed always observed inside nucleated shapes. We now present an experiment demonstrating their stability.

4. Experimental study of shape and texture stability.

Nucleation of Sm-A inside the isotropic phase of the liquid crystalline material gives rise to the so-called Sm-A « bâttonnets » [2]. In figure 8 we have reproduced the original « bâttonnet » drawings by Friedel and Grandjean. From these drawings, it does not seem at once obvious that these « bâttonnets » correspond to the equilibrium shapes that could be expected from our model. To discuss in more detail the agreement with our model, it is convenient to « separate » the stability of the surface shape itself from the stability of the bulk texture for a given surface shape. This will be indeed possible, provided the results of the following experimental study of the « bâttonnet » stability. Because Sm-A are still 1D-crystals, it is not obvious, by analogy with 3D-crystals, that the observed shapes correspond to equilibrium. It is well known for instance that actual crystal facets in nucleated shapes are due to dynamical processes (growth instabilities) than correspond to the ones expected for equilibrium shapes. One cannot either exclude a possible dynamical origin for the focal conics. Moreover, as focal conic textures could be metastable systems, their persistancy in stabilized « bâttonnets » of finite life-time is not a proof of their stability. We now present an experiment which studies the « bâttonnet » stability using an AC electrical field $E$ to act on the bulk focal conic texture of the « bâttonnets ». Spatial field gradients $\nabla E$ also play an important part in controlling the « bâttonnet » locations.

![Typical bâttonnet shapes with their focal conic textures corresponding to the nucleation of Sm-A inside its isotropic phase (from G. Friedel and F. Grandjean [2])](image)

The experimental set-up is the following: we use the compound 10CB (4-n-decyl-4'-cyano-biphenyl) which undergoes a first-order Sm-A → Isotropic phase transition at 50.5 °C. Our cell consists in two horizontal glass plates between which two nickel wires of 50 μm diameter are placed one in front of the other, the extremities being separated by ~200 μm. No other spacers than the wires are used. The wires are connected to a frequency generator delivering a sinusoidal tension in the range 0-200 V and frequency $f$ in the range 0-
10 kHz The cell, filled with the liquid crystal material, is placed inside a temperature controlled oven with 10^{-2} °C temperature resolution. We observe the preparation with a video camera attached to a polarizing microscope. We start, at zero voltage, by observing the isotropic phase. Then we carefully decrease the temperature below the Sm-A \rightarrow Isotropic transition. We observe the appearance of the Sm-A « bâtonnets » nucleating very often on the glass substrates, probably because of the occasional presence of impurities favoring the Sm-A germination. We now follow this procedure: we apply a voltage \( V \sim 100 \) V and variable frequency \( f \). For low frequencies \( f < 500 \) Hz or DC frequencies, we observe an electrohydrodynamical flow probably due to ionic convection. To cancel such effects, we fix \( f \sim 10 \) kHz well above the charge relaxation frequency. We first observe that the « bâtonnets » are more or less attracted towards the nearest electrode. This phenomenon, probably caused by the field gradients, appears as a convenient way to drive the « bâtonnets » out of the glass substrate toward the isotropic liquid. Switching off the electrical field, free floating « bâtonnets » are now easily recognized since they drift slowly inside the isotropic liquid. They closely correspond to the description given by Friedel and Grandjean [2]: they present various external shapes decorated with bulk focal conics having their disclination lines more or less virtual (out of the physical Sm-A volume). They are elongated in a direction which is parallel to the average layer piling direction inside the bulk. This is easily verified by searching for the maximum birefringence darkening between crossed nics. Due to the layer curvature of the focal conics, there is no total extinction. The external « bâtonnet » shapes have more or less the revolution symmetry around the elongation axis, but they often present lateral irregularities, the so-called « saillies » [2], corresponding to local rapid variations of the surface/axis distance. The external shape strongly depends on the « bâtonnet » history: those which unstuck from the glass plates have a free floating shape somehow related to their initial shape. Now, raising the temperature, it is possible to decrease their size down to a few microns and then to let them grow again. In this case, their external shape is more regular and strong « saillies » are absent. However, it often happens that a small « bâtonnet » collapses inside a bigger one, creating a « saillie » localized at the impact level. Usually, such « saillies » take more or less a revolution symmetry after a bulk texture rearrangement. It therefore appears, as will be discussed later, that the external shape is most probably out of equilibrium.

We now isolate a \( \sim 50 \) \( \mu \)m large and \( \sim 100 \) \( \mu \)m long « bâtonnet », containing clearly visible focal conics, between the two electrodes (Fig. 9a) and apply various voltages at a frequency \( f \sim 10 \) kHz. We observe that the « bâtonnet » is oriented by the electrical field in such a way that its long axis becomes parallel to the field lines. However, the main effect of the electrical field is to suppress the focal conic domains. For low tensions, in the range 10-30 V, only the large focal conic domains (with sizes \( \sim 50 \) \( \mu \)m) compared with the « bâtonnet » size are suppressed. Increasing the tension, smaller domains disappear and for a \( \sim 200 \) V voltage, all focal conics disappear (Fig. 9b). The « bâtonnet » presents now a uniformly oriented ideal undistorted bulk (perfect extinction between crossed nics), the normal common to the layers being parallel both to the field lines and the « bâtonnet » axis. In presence of the electrical field, the external surface of the « bâtonnet » is not modified (the surface « saillies » do not relax). However, if the « bâtonnet » had not exactly a revolution symmetry, the latter is « instantly » obtained, i.e. within one video frame (< 20 ms).

Then, to achieve the experiment, we suddenly switch-off the electrical field. The focal conics reappear « instantly », i.e. also within one video frame (< 20 ms), inside the bulk (Fig. 9c). According to our optical resolution, the focal conic texture reproduces a pattern very similar to the previous one.

Before discussing the effect of the electrical field on the bulk texture, which is the important point of the experiment, we first come back to the « saillies » of the « bâtonnets » to
quantitatively discuss their persistance. In a true 3D-liquid, a similar «saillie» of curvature radius ~ R would induce a Laplace pressure \( \delta p \sim \gamma/R \) between the inside volume and the rest of the bulk. This pressure \( \delta p \) would then create a reacting flow relaxing the «saillie» (Fig. 10a). The relaxation life time \( \sim \tau_{3D} \) is defined by the balance between the viscous forces and the pressure forces. Calling \( \eta \) the viscosity, the flow velocity \( \nu \sim R/\tau_{3D} \) is then given by \( \eta \Delta \nu \sim \nabla p \) The spatial derivatives taking place on the same length ~ R, this gives \( \nu \sim \gamma/\eta \), and then

\[
\tau_{3D} \sim \frac{R\eta}{\gamma}.
\]  

(13)

Fig 9. — Vanishing of the focal conic texture inside an Sm-A «bâtonnet» ~ 100 \( \mu \)m long, using a HF electrical field \( E \). The black shapes are the \( \varnothing \) 50 \( \mu \)m nickel wire electrodes a) Before applying \( E \), b) \( E \) is applied, the focal comics all vanish. c) \( E \) is switched off, they «instantly» reappear

Fig 10. — «Saillie» relaxation via the Laplace pressure induced flow (arrows) a) Simple 3D-liquid case b) Sm-A case the permeation phenomenon slows down the relaxation process across the 1D-crystalline direction
For R \sim 10 \, \mu m, \, \eta \sim 0.1 \, cgs and \gamma \sim 0.1 \, cgs, we expect \tau_{3D} \sim 10^{-3} \, s, which corresponds, at our scale, to a very short time (less than a video frame). The problem is that, in the case of smectics, the « saillies » do persist. It was already suggested by Friedel [12] that the smectic ordering limits the flow process. Today, knowing the « permeation » [1] mechanism, this argument can be made more quantitative. The final relaxing flow must pass through the layers (Fig. 10b) and is therefore dominated by the permeation law \nu_\perp = \lambda_p \nabla_\perp \nu. The coefficient \lambda_p is defined [1] through \nu_\perp = D, where D is the auto-diffusion coefficient (this relation is an expression of the « fluctuation-dissipation » theorem). Since, at a molecular scale, translational or rotational diffusions cannot be distinguished, D can be expressed as D \sim K/\eta. This gives \lambda_p \sim \lambda^2/\eta, in which the \lambda penetration length compares again to a few molecular lengths far from second-order phase transitions. We then finally obtain \nu \sim (\lambda^2/\eta)(\gamma/R^2), which gives

\[ \tau_s \sim \left( \frac{R}{\lambda} \right)^2 \frac{R \eta}{\gamma} \sim \left( \frac{R}{\lambda} \right)^2 \tau_{3D}. \] (14)

Numerically, (R/\lambda)^{2} \sim 10^{6} gives \tau_s \sim 10^{3} s \sim 0 \, 3 \, hour. The « saillie » relaxation time is therefore too long to be observed, the life time of the « batonnet » being usually, in our experiment, of order 0.1-1 min. At our scale, the « saillies » are in quasi equilibrium. Note that, however, the « instantaneous » exact revolution symmetry taken by the « saillies » when the strong electrical field is applied, is compatible with the \tau_{3D} « liquid » time, the flow is then inside the 2D-liquid layers.

To explain the effect of the electric field, we build a simplified model: the compound we use (10CB) has an anisotropic electrical susceptibility of magnitude \Delta \varepsilon \sim (+10). The electrical field tends to align the molecules (i.e. the « director » which coincides with the normal to the layers) along the field lines, to minimize the field electrostatic energy. It therefore suppresses the focal conic domains, inducing an ideal bulk with planar, equidistant and parallel layers. In a very simple dimensional model, also dropping all constant coefficients, we associate the following energy

\[ W \sim KL - \Delta \gamma L^2 + \Delta \varepsilon E^2 L^3 + \text{Sat}. \] (15)

with a focal conic domain of size \sim L

The first term corresponds to the order of magnitude of the focal conic bulk distortion energy. The second one describes the order of magnitude of the interfacial energy relaxed by the focal conic. The third one corresponds to the order of magnitude of the bulk dielectrical energy spent by the focal conic in the presence of the electrical field E = V/d. The fourth « saturation » term is required to stabilize the focal conic for the following reason: in the absence of any electrical field (E = 0), the focal conic is stable, i.e. W < 0, for \nu \sim \ell_0 \sim K/\Delta \gamma. Then, as \nu \sim \ell_0 increases, W decreases and the focal conic size would tend to grow indefinitely. In practice, this size is limited either by interactions with the neighbouring focal conics, either by the size of the « batonnet » itself. The « Sat. » term can be visualized as a step function of infinite energy height preventing the focal conic to grow above its actual size. For a non-zero E, the stability of the focal conic disappears for W > 0, i.e. \nu \sim \ell_0 \sim K/\Delta \gamma. Both very large domains and very small domains should then disappear. The vanishing of large domains (L \sim \ell_0) occurs for \Delta \varepsilon E^2 L^3 \sim \Delta \gamma L^2, i.e.

\[ V \sim \left( \frac{K}{\Delta \varepsilon} \right)^{1/2} \frac{d}{(\ell_0 L_0)^{1/2}} \] (16)

With K \sim 10^{-6} \, cgs, \Delta \varepsilon \sim 10, d \sim 200 \, \mu m and \ell_0 \sim 0.1 \, \mu m we find that a 50 \, \mu m large domain
disappears for $V \sim 3 \times 10^{-2} \text{ cgs} (= 10 \text{ V})$ which is in agreement with our experiment. Increasing $V$, $L_\infty$ decreases and smaller domains are cancelled. The total cancellation of all domains is achieved for $V = V_{th}$ when the two roots $L_+$ and $L_-$ coincide. This corresponds to

$$V_{th} \sim \left( \frac{K}{\Delta \varepsilon} \right)^{1/2} \frac{d}{V_0}$$

(17)

and $L_{th} \sim 2 \ell_0$. The same numerical values give $V_{th} \sim 0.6 \text{ cgs} (200 \text{ V})$ which again is in agreement with our experiment. The value $L_{th} \sim 2 \ell_0$ is interesting and means that the smallest focal conic domains are not significantly affected by low fields but are indeed the last to disappear for $V = V_{th} (\sim 200 \text{ V})$.

Let us now resume our experimental results. They concern the stability of both the bulk focal conic texture and the external surface shape of the "bâtonnets". We have measured three characteristics times i) a long $\tau_3$ time ($\tau_3 > \text{ few min} \) corresponding to the existence of the non relaxing «sailles", compatible with our interpretation based on permeation, ii) a liquid-like short time $\tau_{3D}$ ($\tau_{3D} < 20 \text{ ms} \) for the revolution symmetry obtained much probably via an in layer liquid flow, and finally, iii) a short $\tau_{FC}$ time ($\tau_{FC} < 20 \text{ ms} \), for the reappearance of the focal conics as the electrical field is switched off. This experiment shows first that the external surface shape does not correspond to an equilibrium state. Indeed it probably depends rather on the history of the system ("bâtonnets" fusions, eventual growth anisotropies, ). However, the important point of this experiment is that the focal conic texture, which reappears "instantly" as soon as the electrical field is switched off, is not metastable but effectively required for equilibrium. Therefore focal conics appear as stable features of Sm-A "bâtonnets". They correspond to the bulk equilibrium shapes which relax, quasi "instantly", the anisotropic surface energy of external surface shapes, which themselves are most probably out of equilibrium.

5. The general texture relaxing an interface of arbitrary shape.

From the previous experimental results, we are naturally driven to study how to relax an arbitrary given Sm-A/isotropic interface. It has already been mentioned in part 2 that focal conics are conveniently associated with one another if they are limited to cones of revolution, since juxtaposing them along the cone generatrices connects the layers continuously. The cones have then a common apex $O$ and the interstices between them can also be continuously filled with spherical layers centered on $O$ (Fig 5b). Such arrangements are usually used to fill a half space limited by a planar boundary (e.g. glass plate holder). In our case, we wish to fill an Sm-A volume limited by an interface with the isotropic phase of any general shape. Moreover, it must be done in such a way that the focal conic texture sets an orientation as close as possible to the preferred Sm-A layers/interface perpendicular orientation.

Let us consider a general hyperbola($H$)-ellipse($E$) and define a bi-conical domain $B = \{ H, E \}$ by the association of two conical domains sharing the same ellipse $E$ and having their apex on two extremities $H_1$ and $H_2$ of the conjugated hyperbola $H$ (Fig 11). One can define concave or convex bi-cones according to the location of $H_1$ and $H_2$ with respect to the ellipse plane. Now, an important thing to note is that everywhere along the boundary of the bi-conical domain, the Sm-A layers are exactly perpendicular to the surface since the molecules are parallel to the cone generatrices (Fig 11c). This geometrical property gives the clue to the problem associating between them such bi-cones allows us to build more complex surfaces, explicitly the envelope surface of the cones, with which it is possible to approximate a previously given interface and therefore to relax a large amount of its surface energy anisotropy.
Fig 11 — Focal cones limited to bi-conical domains a) Convex bi-conical domain b) Concave bi-conical domain. c) Cross section in a plane containing the ellipse long axis for both convex and concave cases, the layers are everywhere perpendicular to the boundaries.

5.1 GENERAL CONSTRUCTION SCHEME. — Let us now discuss a typical geometrical construction of the focal conic texture for an interface of arbitrary shape. To simplify the discussion, we shall disregard the energies associated with focal cones. This roughly corresponds to assume that the typical lengths associated with the interface curvatures are large compared to the characteristic length $l_0 \sim K/\gamma$. We also disregard the physical processes that created the interface and the "history" of the smectic phase during these processes, we therefore construct the texture ex nihilo.

To build the focal conic texture, we first consider an arbitrary point $O$, inside the smectic bulk, and let issue from $O$ a "bunch" of revolution cones tangent along their generatrices, defining a set of tangent ellipses lying on the interface (Fig. 12a). Each cone can then be transformed into a bi-conical focal conic domain approximating the residual curvature above the ellipse (Fig. 12b). According to the interface curvature sign above each ellipse, the corresponding bi-conical domain can be either convex or concave. The envelope surface of the

Fig 12 — a) An array of cones of revolution, emitted from $O$, define on an arbitrary surface a set of ellipse-like curves, close to which will be located the ellipses of the focal conic domains which reorient the layers at the interface b) The bicones focal domains for convex or concave surfaces.
bi-cone « bunch » constitutes a first approximation of the interface, that can be energetically optimized with respect to the corresponding set of geometrical parameters. Note that this focal conic texture generalizes the particular case of planar Sm-A/isotropic interfaces in which the bi-conical domains are degenerated into cylinders [4] as the point $O$ is removed to the infinite. A good picture of these interfaces can be found in figure 13, which reproduces a drawing from Friedel [12] and a picture of us.

![Diagram](image)

**Fig. 13.** — a) Focal cones at a large SmA-Isotropic interface, according to Friedel. The small radial lines are probably hyperbolae of virtual second generation focal cones b) Corresponding picture of a real interface

### 5.2 SUCCESSIVE FOCAL CONIC GENERATIONS.

In the general case, there remains however, in both concave and convex cases, a gap left between any bi-conical domain $B$ and the interface (see Fig. 12b). Let us first discuss the convex case. If this gap is « very small », i.e. small compared with the « renormalized threshold » as will be discussed later, no further process should go on. Otherwise, to relax more closely the surface energy anisotropy, one can try to refine the envelope approximation in piling smaller focal conic bi-cones inside the gap left above a given ellipse. This can be made with a smaller « bunch » of bi-cones issuing from the apex of $B$ near the interface, both tangent between themselves and with the large ellipse cone below (Fig. 14a). Each new focal conic so generated could also give birth to another generation in the same way (depending on the energetical balance as will be qualitatively discussed later). Such successive generations on Sm-A/isotropic interfaces were first observed by Friedel and Grandjean [2] (Fig. 14b, c). It seems an experimental evidence [2] that the apex $H$ of $B$, in such cases, always lies on the interface. Probably second generations are, in this way, more efficient to approximate the interface closely. Second generations can also be more or less virtual (i.e. out of the physical Sm-A volume) as shown in figure 14a; in this case, only the hyperbola and a part of the ellipse are visible. Qualitative energetical arguments concerning this phenomenon will be given later.

### 5.3 FOCAL CONIC TREES.

The concave case is slightly different. A similar geometrical construction gives rise to the structure shown in figure 15. However, such structures do not seem to have been already observed. Perhaps because the ellipse ($E_1$) of $B$ is much reduced and almost invisible, or perhaps because such a structure would be unstable with respect to
Fig 14 — Second generation focal domains, for a convex surface  a) Since the shape of the surface is not the bicone of the first generation focal conic $E_1 H_1$, a second generation focal domain grows, real as $e_2 h_2$, or semi virtual as $e_2' h_2'$. b) A complete second generation focal conic (from Friedel) c) Semi virtual second generation focal conics (from Friedel) The common focus is virtual, since the curvature distortion is larger there.

![Diagram of second generation focal domains](image)

Fig. 15 — Possible second generation for a concave interface This situation has not yet been observed

![Diagram of concave interface](image)

the sliding of $M_1$ toward $M_2$ (see Fig. 15), building a more equilibrated texture of the first-generation type.

There exists however an interesting structure, very similar to this one, that was not already described by Friedel and Grandjean. It concerns again convex interfaces. We observed it inside a capillary tube in a Sm-A phase growing from an air meniscus interface into the isotropic phase (Fig 16). Instead of nucleating a second focal conic generation inside the gap above a convex bi-conical domain relaxing a convex interface, the smectic sometimes prefers to build arborescent structures. The interfacial apex point of a bi-conical domain
Fig. 16 — Observed focal conic « tree » inside a square section vertical capillary tube of 50 μm. Air/Sm-A interface at the bottom. Sm-A/Isotropic interface above. Three generations of branches are indeed observed.

\([H, E]\) (see Fig. 12) can be drawn back from the interface down to a new position \(H_2\), in order to build now a concave bi-conical domain (Fig. 17a). This defines a cone of revolution \((H_2, E)\) inside which can be placed a new « bunch » of \(N\) convex bi-conical domains \(B_i = [h_i, e_i]\) issuing from \(H_2\), as usual tangent between themselves and the bulk interstices filled with spherical layers centered on \(H_2\). This builds a « focal conic tree » having a hyperbolic « trunk » \(OH_2\), \(N\) hyperbolic « branches » \(h_i\) and \(N\) terminal elliptic « leaves » \(e_i\). Each of the « branches » can give birth to « sub-branches » bearing more elliptic terminal leaves as sketched in the arborescent structure of figure 17b.

The general piling procedure of bi-conical domains must stop when the energetical balance becomes unfavorable. As the conical envelope gets closer to the interface, both the angular discrepancy with respect to the preferred perpendicular orientation decreases and the total number of smaller focal conics increases. The process must then continue down to a « renormalized » size \(\ell_0\) taking these effects into account, and which depends on the particular cases. In the case of asymptotically large equilibrium shapes, an explicit calculation is attempted in the next section. At the last iteration, close to the threshold where the balance

Fig. 17. — Focal conic « trees ». a) General construction scheme b) Extrapolated arborescent texture based on the same principle.
becomes unfavorable, there is no more any reason to place a whole focal conic domain inside the bulk. It then becomes interesting to reorient the surface with partly virtualized focal conics. The virtualization process of the focal conics (toward the isotropic phase) suppresses the regions of highest bulk distortion energy density. This corresponds both to the disclination cores and to the bulk regions close to the point where the hyperbola meets the ellipse plane.

Such half virtualized focal conics are often observed in the vicinity of the interface (Fig. 14c).

We can now qualitatively explain the main characteristics of the Sm-A « bâtonnets »: They have roughly a « Wulf-like » shape, being elongated in such a way that the lateral parts have a low surface energy corresponding to the quasi perpendicular layer/surface orientation. Conversely, the extremities, which correspond to a high surface energy, are conveniently relaxed with a convex bi-conical domain, typically a whole circle-straight line focal conic, as can be seen on Friedel and Grandjean's drawings (Fig 8b). However, relaxing in such a way both ends is problematic because of the association of the two domains. To avoid strains, one should have to place a « radial » point \( O \) in the middle of the bulk where the two conical domains should issue from, and in addition should fill the rest of the bulk either with spherical layers centered on \( O \), either with a corresponding radial « bunch » of domains. In the case of usual « bâtonnets » of size \( \sim 100 \mu \text{m} \), both solutions are probably unfavorable: the first one would set of the orientation of high energy along the lateral parts and the second would instead require the creation of too great a number of focal conics (this is not the case for asymptotically large sizes as will be discussed later in the next section). Sometimes, the sides, more or less in quasi-equilibrium, can be tilted with respect to the « bâtonnet » axis with an angle \( \theta \) (see Figs. 8c-8d for instance), in this case, the layer/interface orientation is not exactly perpendicular and the « renormalized » threshold \( l_0 \) increases In the limit \( \theta \ll 1 \), assuming \( \gamma = \gamma_\perp + \Delta \gamma \sin^2 \theta \), the threshold diverges now as \( l_0^2 \sim K/(\Delta \gamma \theta^2) \). If we require, according to the usual procedure, that the smectic layers should be continuously connected with the central layers perpendicular to the « bâtonnet » axis, we must reorient the surface with focal conic ellipses, the excentricity \( e \) of which is perfectly defined (see Fig. 18); trivial calculations give \( e = \cos \theta \). In addition, the threshold divergence, as already previously explained, gives rise to partly virtualized domains where only a fraction of the hyperbola is real (see Fig. 8d).

The last part concerns the « saillies » (Fig 8c). As discussed in the precedent section, they are probably often created by the radial repartition of the mass of a small « bâtonnet » collapsed

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Fig 18 — Possible « bâtonnet » focal conic in a cylinder-planar domain relaxing an interface tilted by \( \theta \) with respect to the symmetry axis \( \Delta \). Matching with the central layer piling (\( \Delta \) parallel to \( \Delta' \)) fixes the ellipse excentricity \( e = \cos \theta \).
inside a bigger one A « saillie » contains surface parts of high surface energy (oriented perpendicular to the « bâtonnet » axis) These are probably relaxed by the Friedel and Grandjean « festons » [2], which are focal conic arrangements also more or less virtualized (see Figs. 8b-8c). Although the case of « bâtonnets » is difficult to discuss in detail because they are close to the focal conic apparition threshold, the above rules apply qualitatively well.

6. Asymptotic equilibrium shapes.

Let us now come back to true equilibrium, to discuss the case of large Sm-A volumes The equilibrium shape of an Sm-A volume \( \sim L^3 \) nucleated inside the isotropic phase results from a competition between surface energy fixing the external shape and bulk energy fixing the focal conic texture As we have shown, the interesting characteristic of low dimensional crystals such as smectics is that the bulk is the weakest part which in practice adapts its texture of energy proportional to \( L \), to relax the surface energy anisotropy proportional to \( L^2 \), above the typical micronic size \( \ell_0 \sim K/\Delta \gamma \). In the limit \( L \ll \ell_0 \), we have shown that equilibrium configurations should correspond to Wulf-like shapes with no bulk distortions, like in the case of 3D-crystals In the vicinity of \( \ell_0 \), we cannot solve the general equilibrium shape problem, because it corresponds to a true competition between surface and bulk terms This is still typically the case of « bâtonnets » which practical size \( L \sim 10-100 \mu m \) is one or two orders of magnitude above \( \ell_0 \). We can however expect to solve the asymptotic limit \( L \to \infty \), in which the system should present symmetrically a 3D-liquid behavior because of the corresponding bulk infinit weakness.

In the case of a true 3D-liquid, there is no anisotropy and the equilibrium shapes are spheres (smallest surface for a given volume) In the limit \( L \to \infty \), provided we are able to find a bulk focal conic texture relaxing all the surface energy anisotropy, the solution should then be also a sphere. Let us consider an Sm-A sphere \( S \), of radius \( R \) and center \( O \), inside the isotropic phase The sphere being locally plane in the asymptotic limit, it is natural to construct the focal conic texture by analogy with the planar focal conic hexagonal network of reference [4] We consider an arbitrary number of identical focal cones of revolution (circle and conjugated straight line), and place their circles \( C \) on the Sm-A/isotropic interface, tangent between themselves (Fig. 19a) Because of the spherical symmetry, the conjugated straight disclination lines \( H \) are all pointing toward the center Each circle of radius \( \rho \) limits a focal conic convex bi-conical domain of apex \( O \) and base \( C \). The cones being tangent along

Fig 19 — a) Basic focal conic spherical network for asymptotic equilibrium shapes b) « Apollonius » filling of the interstices The sphere is asymptotically relaxed everywhere.
their common generatrices, no strain occurs between contiguous focal conics, as the Sm-A layers match continuously. The interstices can also be continuously filled with spherical layers centered on O. In a first approximation, the surface fraction occupied by the circles has now the perpendicular layer/interface orientation corresponding to the lowest energy $\gamma_\perp$. The total surface energy reads $W_\phi \sim 4 \pi R^2 \{ (1 - \alpha) \gamma_\perp + \alpha \gamma \}$, with $\alpha$ the surface fraction of the interstices. If the relaxation of the surface anisotropy by the focal conics were complete, only a few of them would be present in order to minimize also the bulk distortion energy, as previously discussed in biphased systems [3]. However, the important point for asymptotic shapes is that the relaxation is not perfect, i.e. the Sm-A layers are not exactly perpendicular to the interface. This discrepancy forces the creation of a great number of small focal conics. Quantitatively, the angular mismatch with respect to the perpendicular orientation is of order $\rho/R$, and the integration of the surface energy inside the circles defines a mean value of order $\sim \gamma_\perp + \Delta \gamma (\rho/R)^2$ (the second derivative is of order $\Delta \gamma$ if the function $\gamma(\theta)$ is not too far from a sine square function). In the asymptotic limit $\rho \ll R$, $\alpha$ tends towards a pure constant and the number $N$ of focal conics toward $N \sim 4 \pi (1 - \alpha) R^3/\pi \rho^2$. The bulk energy is therefore $W_\phi \sim NC\rho K\rho + \alpha 8 \pi KR$, in which we have used the generic notation $C\rho = Cte + Cte \ln (\rho/\xi)$, and added the interstitial spherical bulk energy. The total bulk energy can then be written as the sum of a principal term

$$W_\rho \sim (1 - \alpha) 4 \pi R^2 \{ \gamma_\perp + \Delta \gamma \left( \frac{\rho}{R} \right)^2 \} + 4(1 - \alpha) \left( \frac{R}{\rho} \right)^2 C\rho K\rho$$  

and an interstitial term

$$W_i \sim \alpha 4 \pi R^2 \gamma_1 + \alpha 8 \pi KR.$$  

Minimizing $W_\rho + W_i$ with respect to $\rho$ leads to $\rho^3 \sim \ell_0 R^2$. We therefore find that the corresponding solution contains a growing number $N \sim R^{2/3}$ of focal conics whose size also scales as $\rho \sim R^{2/3}$. In the asymptotic limit, the total energy takes the form $W \sim 4 \pi R^2 \{ \gamma_\perp + \alpha \Delta \gamma + O((\ell_0/R)^{2/3}) \}$, and the bulk focal conic energy scaling as $\sim R^{4/3}$ is indeed negligible compared to surface energies.

However, a fraction $\alpha$ of the surface is still not relaxed and this solution is not « optimal ». An optimal configuration must be a sphere (asymptotically) everywhere relaxed in order to have asymptotically the smallest possible energy for any given volume, i.e $W_\infty \rightarrow 4 \pi R^2 \gamma_\perp$. Such a solution can indeed be obtained from the previous one by filling the interstices with smaller focal conics according to an « Apollonius » iterative process (Fig. 19b), as already discussed in planar [5, 9] and similar spherical [3] geometries. Obviously the corresponding straight disclination lines are also pointing towards the center $O$, according to the general focal conic association rules. Let us introduce a second variable $\rho^*$ describing the size of the smallest focal conics filling the interstices, and calculate the new interstitial energy $W_i$. In the asymptotic limit, the sphere can be assimilated locally to a plane, provided $\rho/R = 1$, and we can use the following results [5] on planar Apollonius fillings filling the interstices down to small focal conics of size $\sim \rho^*$ generates a total number of focal conics per interstice which scales as $M(\rho^*) \sim (\rho/\rho^*)^n$, in which $n$, the Apollonius scaling index, in the planar case, is close to $n \sim 4/3$. Trivial differential calculations show that the total generated perimeter of circles scales as $P(\rho^*) \sim M(\rho^*) \rho^*$ and that the resulting unfilled surface left per interstice scales as $S(\rho^*) \sim M(\rho^*) \rho^*$. Using a similar calculation, it can be shown that the total interstitial integrated bulk focal energy down to $\rho^*$ also takes, in spite of the logarithmic dependence, the form $\sim M(\rho^*) C\rho^* K\rho^*$. In the asymptotic limit, the total number of interstices is close to $2 N \sim (R/\rho)^2$, since the great circles build a quasi hexagonal
pattern, and the unrelaxed fraction of the total interstitial surface around the sphere scales therefore as \( \varepsilon(\rho^*) \sim (\rho/\rho^*)^{n-2} \). The new interstitial energy is now a function of the two independent variables \( \rho \) and \( \rho^* \) \((R > \rho > \rho^*)\), and now reads

\[
W_i' \sim \alpha 4 \pi R^2 \left\{ \varepsilon \gamma_\parallel + (1 - \varepsilon) \gamma_\perp \right\} + 2 NM C_p \, K \rho^* + \alpha \varepsilon 8 \pi KR
\]  
(20)

in which we have neglected the small angular mismatch with respect to perpendicular orientation inside the interstitial focal conics \((\gamma \sim \gamma_\perp)\). \(W_i'\) can be more explicitly rewritten, introducing the scaling index \( n \) and dropping now all constant coefficients, as

\[
W_i'' \sim R^2 \left\{ \gamma_\perp + \Delta \gamma \left( \frac{\rho}{\rho^*} \right)^{n-2} \right\} + \left( \frac{R}{\rho} \right)^2 \left( \frac{\rho}{\rho^*} \right)^n C_p \, K \rho^* + \left( \frac{\rho}{\rho^*} \right)^{n-2} KR
\]  
(21)

We can now minimize the sum \( W = W_p + W_i' \) with respect to the two independent variables \( \rho \) and \( \rho^* \). In (21), the third term is negligible (compared to the second term inside the brackets) in our limit \( R \gg \ell_0 \). Minimizing with respect to \( \rho^* \) leads

\[
\rho^* \sim \ell_0^*.
\]  
(22)

A similar result was already obtained in biphased systems [3]. This result is indeed simple to understand, since it is in fact independent of the Apollonius piling. It is an individual property that any intermediate focal conic, whose radius \( \rho_1 \) is larger than \( \ell_0 \), is energetically favored, i.e. \( \delta W \sim C_p, K \rho, -\Delta \gamma \pi \rho \pi^2 \ll 0 \) for \( \rho_1 > \ell_0 \). Less trivial is the reaction on the behavior of the principal hexagonal focal conic network, as we show now. The interstitial energy can be rewritten as

\[
W_i' \sim \alpha 4 \pi R^2 \left\{ \gamma_\parallel + \text{Cte} \Delta \gamma \left( \frac{\rho}{\ell_0} \right)^{n-2} \right\}
\]  
(23)

and the sum \( W = W_p + W_i' \) becomes

\[
W \sim 4 \pi R^2 \left\{ \gamma_\parallel + \text{Cte} \Delta \gamma \left( \frac{\rho}{R} \right)^2 + \text{Cte} \Delta \gamma \left( \frac{\ell_0}{\rho} \right)^{2-n} + C_p K \right\}.
\]  
(24)

In the asymptotic limit \( L \to \infty \), since \( 2 - n \sim 0.7 \) is strictly less than unity, the fourth term corresponding to the bulk energy of the principal focal conics \( \sim \rho \) is negligible with respect to the third one describing the total interstitial energy. The competition which defines the asymptotic equilibrium value of \( \rho \) is now between the second and third terms, i.e. between the efficiency of the great focal conics, which requires \( \rho \ll R \), and the efficiency of the interstitial filling requiring \( \rho \gg \ell_0 \). Minimizing \( W \) with respect to \( \rho \) yields \( \rho^{4-n} \sim \ell_0^{2-n} R^2 \) and we find that \( \rho \) now scales as

\[
\rho \sim R^{2/(4-n)}
\]  
(25)

With \( n \sim 4/3 \) we find \( 2/(4-n) \sim 0.75 \). Because of the gain due to the interstitial filling, the principal focal conics can increase in size. As for the total bulk energy associated with all focal conics, the interstitial term is now the leading one and scales as \( R^{1+m} \), with \( 0 < m < 1 \) given by \( m = n/(4-n) \sim 0.5 \). This solution is now indeed optimal since the total energy behaves as

\[
W_\infty \sim 4 \pi R^2 \left\{ \gamma_\parallel + O \left( \left( \frac{\ell_0}{R} \right)^{1-m} \right) \right\}
\]  
(26)
since the interstices are now also relaxed. It asymptotically coincides with the energy of the equilibrium shape of a true isotropic 3D-liquid with $\gamma_{\text{iso}} = \gamma_{\perp}$.

To describe the bulk texture of this asymptotic solution more precisely, we must examine which distortion is left near the sphere center. As already briefly recalled in part 2, the core of a focal conic straight disclination line must « vanish » far from the circle, i.e. near the center in our case. It was predicted [11] that the angular discontinuity of order $\theta \sim \rho/h \ll 1$ at a large distance $h$ from the circle along the hyperbola line must transform into a smooth elastic connection of larger radius $r_c \sim \xi/\theta$, involving a smaller energy of order $K\theta^2$ per unit length. Therefore, although the line energy becomes indeed negligible for $h \gg \rho$, the core still remains quasi microscopic on a longer distance. Let us determinate how deep it does penetrate in our case: this corresponds to a length $h_p$ for which the « exploded » core of radius $r_c \sim \xi(h_p/\rho)$ compares with the local radius of the conical domain containing the focal conic, i.e. $\xi(h_p/\rho) \sim \rho (1 - h_p/R)$ which gives

$$h_p \sim \frac{R}{1 + \frac{R^2}{\rho^2}} \sim \frac{R}{1 + \frac{\xi}{\ell_0} \left( \frac{\ell_0}{R} \right)^m} \sim R$$

(27)

Since equation (25) reads explicitly $\rho^2 \sim \ell_0^{1-m} R^{1+m}$ With $m \sim 0.5$, we find that $h_p$ compares with $R$. This means that the straight lines do not « physically » vanish but do penetrate down to the center, although with an energy negligible compared with the energy of the whole focal conic.

Finally, we have shown that asymptotic optimal shapes are indeed possible. However, there may exist solutions slightly different from that previously described, although having the same asymptotic energy. For instance, to relax the energy due to the small angular discrepancy inside the circles of the principal focal conics, one could imagine planar cuts as discs laying on the focal conic circles. This would result in an increase of the total surface with respect to the sphere for the same volume, corresponding to an energy term equivalent to that previously discussed (first term of Eq (18)). Note that such a configuration is not really « faceted », since it is asymptotically equivalent to a true sphere. One could also imagine other ways to relax the energy due to this angular discrepancy, e.g. second focal conic generations inside the interstices above the principal circles, focal conic arborescent structures, etc... However, all these derived optimal configurations look roughly similar and the choice between them depends probably on the details of the surface energy function.

Our precedent qualitative arguments concerning the necessity of focal conics inside equilibrium shapes were based on the competition between surface energies scaling quadratically in length and focal conic energies assumed to scale linearly. This indeed is locally true for each focal conic. However, inside a given equilibrium shape, a great number of focal conics appear and the corresponding total energy then scales according to a power law whose exponent becomes $1 + m$. Obviously, we shall always find $1 < 1 + m < 2$ as illustrated for instance by the previous asymptotic solution giving $m \sim 0.5$. From equation (26) we deduce that optimal solutions with $m$ close to zero are energetically the most favored. Now, the Appolonius piling is a self similar process. In the asymptotic limit of large sizes, the hierarchy of focal conics constitutes a fractal object of dimension $1 + m$. Note that $m$ is simply relaxing the surface energy anisotropy by analogy with 3D-crystals facets. One focal conic replaces locally an unfavourable Sm-A/Isotropic interface of dimension 2, by an unfavourable Sm-A/Isotropic interface, the core of the disclination ellipse and hyperbola, of dimension 1. The total relaxation of a large surface by an Appolonius packing of focal conics replaces the unfavourable interface of dimension 2 by that of the quasi fractal array of focal conics of dimension $1 + m \sim 1.5$, which remains anyway smaller than 2.
To complete this study, more work remains to be done. First, still about Sm-A/Isotropic interface, one could try to describe the exact shape of «bâtonnets», in more detail by predicting their exact equilibrium shape and texture in presence of focal conics. In fact, this seems very delicate, since it is quite difficult to grow and stabilize isolated individual «bâtonnets». A more complicated problem remains the one of filling space with focal conics. We know how to construct a focal conic array around a single centre, from which issue bicone bunches. What happens if we use two or more centers? Is it possible to fill any arbitrary volume with the smectic material, such that its layer remains normal to the Sm-A/Isotropic interface?

We could also try to change the angular anchoring condition of smectic layers to explain other possible interfacial anisotropies with nematic phase, or other materials. In principle, oblique anchoring is possible. One can always cut focal conic texture to yield constant oblique orientation of smectic layers at the interface. The problem is now that one loses the property of easy contact along generatrices of the focal conic domains with perpendicular anchoring.

As for the simple problem of spontaneous plane paving, this could induce an additional frustration which would make the bulk faceting less efficient. Finally, we have only discussed the problem of free surface shapes. In the case where the shape of the related to \(n\), and therefore to the Hausdorff dimension \(2 - n\), somehow describing the «porosity» of the fractal filling. Let us finally note the following analogy: the core of the focal conics, which has no more smectic ordering, can be considered as an «isotropic» line, where the smectic layers are almost in the parallel unfavourable situation. The bulk decoration of the sphere can be visualized as a change of dimension of the unfavourable Sm-A/isotropic interface from 2, the sphere area, to \(1 + m\), the fractal Appolonius array of focal conics.

7. Conclusion: bulk faceting.

In summary, we have studied how the anisotropic surface energy of Sm-A/isotropic «intrinsic» interfaces determines the surface shape and bulk texture of Sm-A volumes inside the isotropic phase. The main result is that «intrinsic focal conics» are induced inside the bulk to set the preferred perpendicular layers/surface orientation. This occurs above a characteristic size of Sm-A droplets \(l_0 \sim K/\Delta \gamma\) of order 1 \(\mu\)m for usual thermotropic Sm-A. This length compares, for an Sm-A volume of size \(\sim L\), the energy of the focal conic texture \(\sim KL\), with the anisotropy of the surface energy \(\sim \Delta \gamma L^2\). To discuss equilibrium shapes, we have studied three regimes according to the magnitude of \(L\) with respect to \(l_0\). Both cases \(L \ll l_0\) and \(L \gg l_0(L \to \infty)\) have been quantitatively discussed. For \(L < l_0\), the Sm-A should present a 3D-crystal behavior and an equilibrium elongated shape given by the Wulff's construction, therefore with an «ideal» undistorted bulk. Conversely, in the limit \(L \gg l_0(L \to \infty)\), asymptotically large equilibrium shapes should present an antagonistic 3D-liquid behavior. In such a case, the bulk rigidity can be considered as infinitely «weak» and should adapt its texture in radial focal conic networks relaxing all the surface energy anisotropy on a spherical liquid-like shape. The case \(L \sim l_0\) is much more delicate since bulk texture and surface shape are in true competition. Experimentally, the «bâtonnet» nucleated shapes, of sizes in the range 1-100 \(\mu\)m not far from the threshold \(l_0\) do contain focal conics. We have experimentally demonstrated, acting on the bulk focal conics of the «bâtonnets» with an electrical field, that it constitutes a true equilibrium texture. Conversely, our experiment indicates that the external shape is out of equilibrium, as is usually the case for 3D-crystals (Smectics are still 1D-crystals). Although often largely virtualized because of the proximity to the threshold, the focal conics appear inside the «bâtonnets» at such places where they are required to relax the surface energy anisotropy for each given external shape.

We have discussed the general method to relax the surface anisotropic energy of a large
interface of any general shape. Our model explains most of the observed textures near Sm-A/isotropic interfaces: paving with tangent focal conic ellipses, further generations, focal conic trees, etc. The general method consists in packing together bi-conical focal conic domains in order to approximate the interface with the *envelope* of these cones. Each bi-conical domain has exactly everywhere along its boundary the favored perpendicular orientation. In this sense, it can be geometrically considered as a « bulk facet » by analogy with 3D-crystal facets which are also surface domains of lowest energy. Although the facets can be determined by Wulff’s construction, there still does not exist any corresponding construction for smectic textures. Still by analogy with faceting, further focal conic generations and smaller interstitial focal conics can be compared with smaller facets of higher sets of Miller indices, and also the spherical filling between bi-conical domains is equivalent to the smooth curved surface between facets.

Finally, focal conics can be considered as *intrinsic* defects of Sm-A phases in contact with their *isotropic* phase, as « bulk facets » smectic interface is imposed, as for instance in a more or less constrained sample between to parallel glass plates, *extrinsic* focal conics appear. Some are created by just antagonistic anchoring, others appear as a memory of the smectic growth process. It is an important practical problem to get rid of these defects by a carefully controlled growth process. In this respect, the understanding that smectic-isotropic interfaces must present their own intrinsic focal conics may be of some importance to grow smectic single crystals.

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Erratum

Evidence for a jacketed nematic polymer

F. Hardouin, S. Mery, M.F. Achard, L. Noirez and P. Keller

(J. Phys. II France 1, n°5 (1991) 511)

The figure 3 is as follows:

Fig. 3 - Quadratic sizes in directions respectively parallel (R//) and perpendicular (R\textperp) to the magnetic field as a function of temperature: I: isotropic liquid, N: nematic.

instead of:

which corresponds to the figure 10:

Fig. 10 - Thermal evolution of the average radius of gyration in the nematic range.