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J. Fournier, Geoffroy Durand. VOLUMIC FACETING OF SMECTIC-A LIQUID CRYSTALS. Journal de Physique Colloques, 1990, 51 (C7), pp.C7-157-C7-162. 10.1051/jphyscol:1990715 . jpa-00231114

HAL Id: jpa-00231114

<https://hal.science/jpa-00231114>

Submitted on 4 Feb 2008

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VOLUMIC FACETING OF SMECTIC-A LIQUID CRYSTALS

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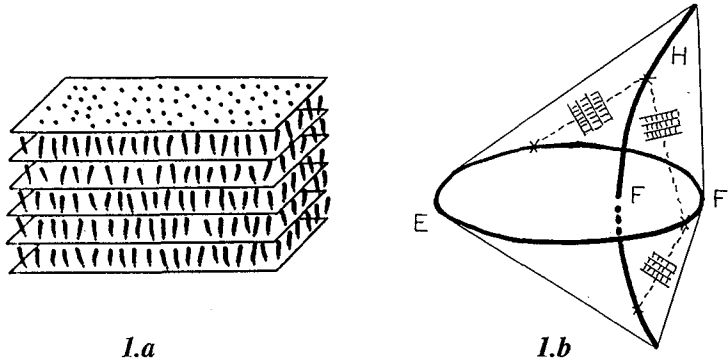
RESUME - Les coniques focales qui décorent les interfaces Smectique-A/ Isotrope sont des défauts de volume intrinsèques qui relaxent l'anisotropie des énergies d'interface. Elle réalisent, pour les systèmes lamellaires un véritable facetage en volume, analogue au facetage de surface des cristaux usuels à trois dimensions.

ABSTRACT - The focal conics decorating the Smectic-A/ Isotropic interfaces are intrinsic bulk defects. They relax the surface energy anisotropy, realizing for lamellar systems a volumic faceting analogous to the surface faceting of usual three dimensional crystals.

I) INTRODUCTION

Smectic-A (*Sm-A*) are lamellar liquid crystalline phases made by the piling of two-dimensional (*2D*) liquid layers. Inside each layer, rod-like molecules are oriented with their long axis normal to the lamellae. *Sm-A* phases are then both *1D-crystals* and *2D-liquids* (Fig.1a). In 1910, G.Friedel and F.Grandjean observed *Sm-A* drops laying on a glass substrate^[1], with a polarizing optical microscope. Such drops did not show "ideal" *Sm-A* phases with parallel equidistant planar layers, but regular networks of defects. These defects, called "Focal Conics" (*FC*)^[2] are now well-known and characteristic of lamellar phases. The *FC* structure consists in bulk domains in which the *Sm-A* layers are curved at *constant thickness*, because lamellae curvature involves much less distortion energy than layer dilation^[3]. The curvature energy density scales like $\sim K/L^2$, K being a traditional elastic constant ($K \sim 10^{-6}$ cgs) and L a typical curvature radius; the *FC* bulk curvature energy scales then as $\sim KL$, L being the size of the *FC* domain^[4]. Such a structure requires topological singularities ("disclinations") where the *Sm-A* locally melts. Obviously their energy should not exceed the total bulk distortion energy; therefore, with our

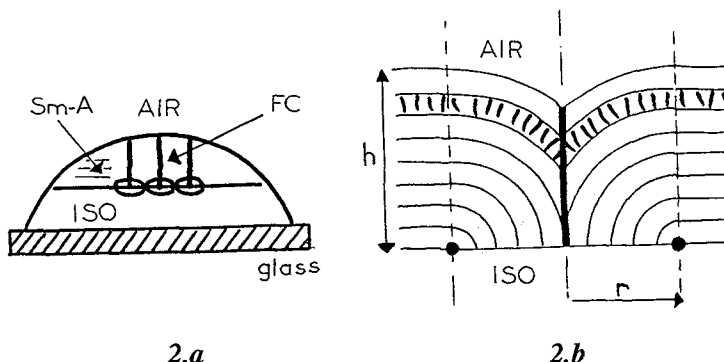
previous argument, only *line* singularities are acceptable (*not* surfaces). In the most general case, the disclination lines are "confocal" *ellipses* and conjugated *hyperbolae* (Fig. 1b).



FCs are quite always present in *Sm-A* samples. They are traditionally explained by substrate induced antagonistic "anchoring directions" of the *Sm-A* layers, forcing layer curvature. Note that, long before X-rays structural studies, it is from the very observation of confocal ellipses and hyperbolae visible *FCs* defect lines that G.Friedel and F.Grandjean first deduced the layered structure of *Sm-A* phases. From then, the geometrical features of *FC* domains have been deeply studied^[5] but many physical problems concerning their stability and their origin are still unsolved.

II) THE *Sm-A* PLATE INSTABILITY

Investigation of processes occurring during *FC* nucleation requires dynamical studies and attention to interfacial phenomena. From this point of view, we have reproduced^[6] in Orsay the old *Sm-A* drop experiment of G.Friedel and F.Grandjean who originally obtained *FC* networks after cooling an Isotropic(*Iso*) droplet toward the *Sm-A* phase. We have controlled the thickness of the *Sm-A* plate growing from the top air interface of the *Iso* droplet with a well-defined thermal gradient. The *FC* networks does appear when the *Sm-A* plate is *still in contact with the Iso phase* (Fig. 2a) and *disappears* at contact with the glass substrate when treated ("silane" coating) to induce *parallel* layer/substrate orientation. These interfacial floating networks are energetically stable. The *Sm-A* plate thickness is the reversible control parameter fixing the size $r(h)$ of the *FCs*. For low thicknesses, the stable state is a monocristalline plate without defects, the *Sm-A* layers being *parallel* to both interfaces; above a thickness threshold, the *FC* network is created by a first-order textural instability.



The density drop at the air interface forces the Sm-A layers to grow *parallel* to the interface^[7]. Then, instead of the assumed antagonistic smectic layer anchoring on the glass substrate, we introduced an *anisotropy* $\Delta\gamma = \gamma_{//} - \gamma_{\perp}$, of the Sm-A/Iso interfacial energy favoring the orientation of the layers *perpendicular* to this interface and inducing the FC network curvature (Fig.2b). A FC of radius r ($r \sim h$) relaxes a surface energy $\sim \Delta\gamma r^2$, but implies a curvature energy $\sim Kr$ and is therefore *stable* above a typical size $l_0 \sim K/\Delta\gamma$. A precise model giving the thresholds and the law $r(h)$ is given in Ref.6 and leads to $\Delta\gamma \sim 0,05$ cgs and $l_0 \sim 0,1-1$ μm . Note that a similar characteristic length was already introduced^[8] in another context.

III) FOCAL CONICS AS VOLUMIC FACETS

The previous experiment has shown that not only external solid substrates but also "natural boundary" interfacial anisotropies, such as the Sm-A/Iso one, can induce FCs. It is of some interest to compare this phenomenon, which seems general for lamellar systems, with 3D-crystals *faceting*, i.e: What would be the equilibrium shape of a Sm-A closed volume having nucleated inside the Iso phase? We keep assuming the minimal surface energy to be reached for layers *perpendicular* to the interface. If one forbids bulk distortions, as for 3D-crystals, the equilibrium shape is obtained by minimizing the interfacial energy for a fixed total volume, i.e. by solving the Wulf's problem^[9]

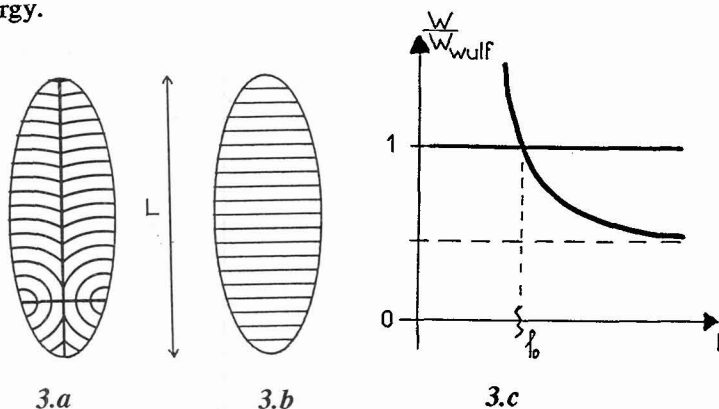
$$\oint \gamma dS + \alpha V = \min \quad (1)$$

with the implicit assumption that the crystalline volume ordering will not change in the process. For a given γ function, the solution corresponds to a defined *homothetical* shape (Fig. 3a) given by "Wulf's construction" in which the relative surface areas of low energy increase. Note that facets are due to discontinuities in the derivatives of γ near orientations of *low* Miller indices (crystallines planes). In the case of smectics which are *soft* 1D-crystals, a volume contribution describing the

free energy f_d associated with all possible volume distortions^[3] must be added in Eq.1, so that the generalized Wulf's problem now reads :

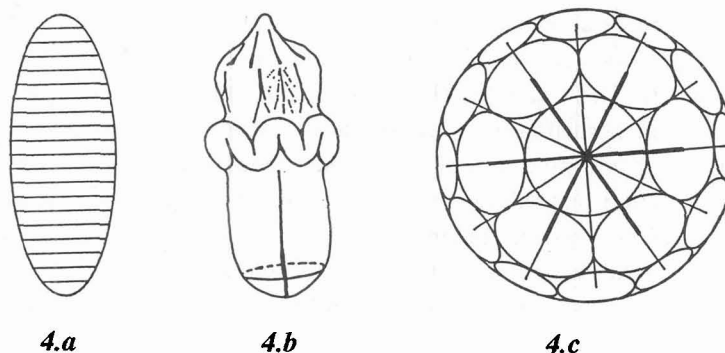
$$\oint \gamma dS + \int f_d dV + \alpha V = \min \quad (2)$$

As for FCs, it can be shown that only curvature energy plays an important role for macroscopic sizes. However, mathematical solutions of *this* new problem are unknown. We will show that solutions of much *lower energy* than Wulf's one can be obtained if bulk FCs are included. From the homothetical property, the energy of Wulf's solution (Fig. 3b) scales like $W_w \sim \alpha \gamma L^2$, in which γ is a mean value defined by the integration of the surface energy. Placing a "revolution" FC (circle-straight line - Fig. 3a) of homothetical radius $R \ll L$ sets a better layer/surface orientation, closer to the perpendicular one, on one end. It reduces the surface energy down to $W_s \sim \alpha \gamma_{FC} L^2$, with $\gamma_{FC} < \gamma$. Since the FC energy $\sim KL$ is negligible compared to surface terms $\sim L^2$, this solution is better than Wulf's one for $L \gg l_0$ (Fig. 3c). FCs should then be present in equilibrium shapes above the size l_0 since they relax a finite part of the surface energy.

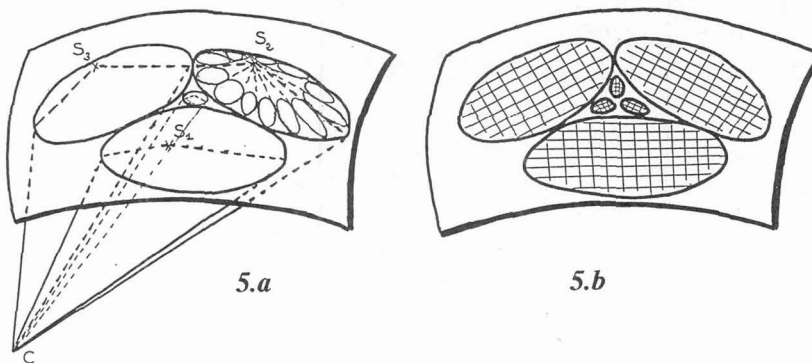


We can distinguish three regimes : *i*) $L < l_0$, Wulf's solution is valid since FCs involve too high energies (Fig. 4a). For usual thermotropic *Sm-A*, l_0 is quasi sub-micronic and then hardly visible. *ii*) $L > l_0$: A mixed "Wulf's type" surface plus bulk FCs should correspond to equilibrium. Real "batonnets" (see Friedel^[2] drawings sketched on Fig. 4b) may have external surfaces out of equilibrium resulting from dynamical growth processes, but are well explained by our model. The FC distribution inside real "batonnets" can be *erased* with an aligning electrical field and instantly *reappears* when the field is switched off. FCs do then correspond to equilibrium states. *iii*) $L \rightarrow \infty$; In that limit, we can predict an interesting asymptotic shape in which the surface orientation is *everywhere optimal*. The external surface must then be a sphere with a radial FC network relaxing all the

anisotropic part of γ (Fig. 4c). Such a solution has indeed an optimal asymptotic energy $W \rightarrow 4\pi L^2 \gamma_{\min}$, L been the radius of the sphere and $\gamma_{\min} \equiv \gamma_{\perp}$. The existence of these asymptotic shapes could be studied in low gravity experiments.



In the limit of large sizes, a *Sm-A/iso* interface of *any given shape* can be relaxed by an adequate *FC* distribution (Fig. 5a) with the ellipses lying *on* the interface. To avoid elastic strains between *FCs*, *FCs* must be limited to *bi-conical domains* (Fig. 1b), the neighbouring domains being tangent along the generatrices of the cones^[5]. *FC* bi-conical domains have the interesting property that the *Sm-A* layers are everywhere *perpendicular* to the boundary, which we know corresponds to the minimal *Sm-A/iso* surface energy. Therefore such *FC* bi-conical domains appear as individual "bricks" of *lowest surface energy* for the bulk texture construction. This is qualitatively analogous to the construction of a *3D*-crystal surface equilibrium shape with *facets* being the individual bricks of *lowest energy* (Fig. 5b). One often observes "second *FC* generations"^[2] (right part of Fig. 5a) and interstitial smaller *FCs*. This would correspond to the appearance of smaller facets of higher Miller indices sets as also sketched on Fig.5b. The *smooth* parts between facets would correspond to the assumed *interstitial spherical filling*^[10] between biconical *FC* domains. It follows that *FCs* are intrinsic defects of *Sm-A* in contact with their isotropic phase, the *FC* bi-conical domains playing the role of "volumic facets" for equilibrium shapes.



IV) CONCLUSION

The plate instability was created by *two* antagonistic parallel/perpendicular favored orientations. More generally, the *unique* perpendicular favored orientation of the *Sm-A*/Isotropic interface alone creates also topological constraints and leads, as previously shown, to *volumic faceting* by *FC* nucleation. This constraint in our euclidian R_3 space could be released in a *curved* space, as recently demonstrated in a *2D*-model[11]. A similar *3D*-study remains to be done. *FC* appearance is a more general phenomenon in *Sm-A*, as shown for instance by the internal texture of *Sm-A* droplets inside mixtures of *Sm-A*-amorphous liquids[2]. However, it has not been demonstrated that the anisotropy of these other Isotropic-smectic interfaces is here the leading creation mechanism. Another example is the one of *short* pitch "cholesterics" which do show *FC*s and behave as *stiff* lamellar phases. Finally, the possibility of surface energy minimization by volume curvature applies also to tubular systems (i.e. *1D*-liquids and *2D*-crystals) where non-compressive curvature is restrained to one direction. It would be interesting to check experimentally if the "developpable domains"[12] of discotics, for instance, are intrinsic defects or not of the *Iso*/discotic interface.

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