A coarse-grained theory of wrinkle patterns induced by geometric incompatibility

The smectic order of wrinkles
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Nature Communications **8**, 15809 (2017)

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The frustration that stems from Gauss' *Theorema Egregium* is familiar to anybody who attempted to draw a map of earth or to neatly cover a ball with a wrapping paper. This fundamental theorem asserts that it is impossible to deliver *isometrically* (*i.e.* with no in-plane strain) a two-dimensional solid with "target" Gaussian curvature, $G_{\rm tar}$ (defined by its intrinsic metric), to the surface of a "substrate" with Gaussian curvature, $G_{\rm sub} \neq G_{\rm tar}$. Figure 1 illustrates the implication of this "geometric incompatibility" for thin solid objects, whose thickness, t, is much smaller than their diameter, D: **(a)** Top view of a solid sheet "stamped" in a narrow gap, δ, between two rigid spheres of radius R [1] (here, $G_{\rm tar}$=0, $G_{\rm sub}$= R^{-2}); **(b-d)** Top views of polygonal and circular patches cut from a spherical shell with radius of curvature R, floating on a flat liquid surface [2] (here, $G_{\rm tar}$=R^{-2}, $G_{\rm sub}$=0); **(e)** Top view of a cone confined by rigid plates ($G_{\rm tar}(\mathbf{x})$=$\alpha\cdot\delta_{2d}(\mathbf{x})$, $G_{\rm sub}$=0, α is the cone's angle).

The origin of wrinkle patterns in the above examples can be understood by recalling the thickness-dependent ratio, $B/Y \sim t^2$, between the bending modulus (B) and stretching modulus (Y) of a thin Hookean solid object, and considering the respective energies (per area), associated with the corresponding deformation of the solid, as well as the penalty due to deflections from the (incompatible) confining topography:

$$u_{\rm strain} \sim Y \cdot ({\rm strain})^2 \;\; ; \;\; u_{\rm bend} \sim B \cdot ({\rm curvature})^2 \;\; ; \;\; u_{\rm sub} \sim K \cdot ({\rm deflection})^2 \; . \qquad (1)$$

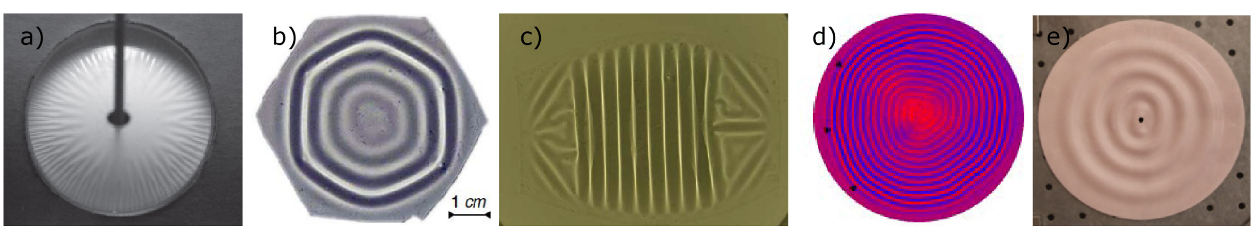

Figure 1: (a) spherical stamping of a solid sheet [1]. (b-c) experiment and (d) simulation of various patches of a spherical shell floating on a liquid surface [2]. (e) cone stamped between flat plates (courtesy of E. Sharon).

Here, K is a "substrate stiffness" parameter, *e.g.* in Fig. 1b, $K = \rho_{liq} g$, due to gravity that acts on the liquid foundation. In stamping problems, the energy $u_{\rm sub}$ is replaced by a *non-holonomic* constraint (*i.e.* deflection in Figs. 1a,1e is limited by the gap δ).

If the confinement is sufficiently strong (*i.e.* $K \to \infty$ in Figs. 1b-d ; $\delta \to t$ in Fig. 1a,1e), the solid object conforms perfectly to the substrate and is highly strained. However, if the confinement is imposed more softly, compression may be relieved by deflections from the substrate, thereby "trading" an energetically-expensive strain with energetically-cheap curvature. This mechanism underlies the formation of wrinkles with characteristic wavelength, $\lambda \sim \delta$, in stamping problems (Fig. 1a,1e), or $\lambda \sim (\frac{B}{K})^{1/4}$ (Fig. 1b-d), reflecting a balance between $u_{\rm bend}$, which favors small curvature (hence large λ), and $u_{\rm sub}$, which favors small deflections (hence small λ). In uniaxial compression of a solid sheet, where there is no geometric incompatibility, uniformly-spaced, parallel wrinkles suppress the "*bare*" strain, $\varepsilon_{\rm bare}$ (reached at $K \to \infty$) down to a "*residual*" value, $\varepsilon_{\rm res} \sim \frac{\sqrt{BK}}{Y}$ [4]. In a geometrically-incompatible confinement (Fig. 1), the wrinkle patterns are often far more complex, and became a focus of attention in the "extreme mechanics" community. Since the energetic price tag for strain is very large, a natural conjecture is that minimization of the residual strain, $\varepsilon_{\rm res}$, selects among various wrinkle states that accommodate the geometric confinement. A central object of study is thus the ratio $\frac{\varepsilon_{\rm res}}{\varepsilon_{\rm bare}}$ (where $\varepsilon_{\rm bare} \sim |G_{\rm tar} - G_{\rm sub}| \cdot D^2$). Does this ratio approach a finite value as the confined solid is made infinitely thin? Alternatively, are there "*asymptotically isometric*" deformations [3] for which this ratio vanishes in that limit?

In a recent study [2], Aharoni *et al.* laid out an innovative approach to geometrically-incompatible confinement. In their experiments and simulations, the authors cut patches of various shapes from a thin spherical shell, which were then placed on the flat surface of a strongly adhesive liquid (Fig. 1b-d). A prominent motif (see Figs. 1b-c) is the emergence of *wrinkle domains*, in which the pattern appears to relieve a uniaxial compression through parallel, uniformly spaced, elongated wrinkles. Drawing an analogy to liquid crystals with a smectic order, whereby molecules form a layered structure [5], the authors proposed to describe domain properties through an *effective 2D smectic* energy. In this coarse-grained description, the smectic "compression" and "bending" modulii reflect the energetic penalties associated with deviations of the average wrinkle wavelength from the energetically-favorable value, $\lambda = (\frac{B}{K})^{1/4}$, and deviation of the average wrinkle direction from the compression direction. These effective smectic modulii should not be confused with Y and B in the original energy (Eq. 1); instead, they are determined through nontrivial combinations of all physical parameters: the "bare" elastic modulli, Y, B, the stiffness K, and the bare strain, $\varepsilon_{\rm bare} = (\frac{D}{R})^2$.

Armed with their coarse-grained theory, the authors pushed forward to obtain a number of powerful results. Employing the vast literature on domain walls (grain boundaries) in liquid crystals, they provided nontrivial, quantitative predictions for the width and shape of the transition zones between wrinkle domains, in terms of the control parameters, Y, B, K, and $\varepsilon_{\rm bare}$. Another notable result pertains to long wavelength undulations (Fig. 2d), predicted through analogy to a well-known, boundary-induced instability in smectic phases. Finally, "polygonizing" a spherical shell into locally-planar domains of size $L_{\rm dom}$ contributes to the residual strain, which can be estimated as: $\varepsilon_{\rm res} \sim (\frac{L_{\rm dom}}{R})^2 + \frac{\sqrt{BK}}{Y}$ (the second, "mechanical" term, exists even in uniaxial compression [4]). Balancing the domain-bulk energy, $Y \cdot {\varepsilon_{\rm res}}^2$, with the domain-boundary energy (which favors $L_{\rm dom}\to D$), a novel scaling rule is obtained for the domain size: $\frac{L_{\rm dom}}{D}\sim(\frac{R\cdot t}{D^2})^{1/5}$. Notably, for sufficiently thin shells ($\frac{t}{D}\ll 1$), the ratio $\frac{\varepsilon_{\rm res}}{\varepsilon_{\rm bare}}$, and the number of domains, $N_{\rm dom}\sim(\frac{D}{L_{\rm dom}})^2$, are determined by the ratio, $\frac{D}{\sqrt{R\cdot t}}$, between the solid's diameter and the geometric mean of the largest and smallest scales (R, t, respectively), recognized as a "*geometric bendability*" parameter [3]. The independence on the strength with which confinement is imposed (*i.e.* K), suggests that these scalings characterize also a stamping version of this problem (a spherical shell confined between planar plates).

Putting together these results, Ref. [2] offers an appealing, multi-scale plan of attack. At scales smaller than the wrinkle wavelength λ, the mechanics is reminiscent of a planar sheet under uniaxial compression; at intermediate scales (between λ and $L_{\rm dom}$), the solid responds as a 2D smectic matter; at scales larger than the domain size, the mechanics is finally described by elastic solid theory (Eq. 1), albeit with renormalized modulii. In order to assess to value of this approach for analyzing morphologies of geometrically-confined solids, it is crucial to address a few potential difficulties.

• For a given, axially-symmetric confinement problem, characterized by the shell's radius of curvature (R) and diameter (D), the analysis of [2] shows that for sufficiently thin shells ($\frac{t}{D}\lesssim\frac{D}{R}$), a wrinkle domain pattern is preferable to a planar, unwrinkled deformation, and becomes asymptotically isometric (*i.e.* $\frac{\varepsilon_{\rm res}}{\varepsilon_{\rm bare}}\to 0$) as $\frac{D}{\sqrt{R\cdot t}}\to\infty$. However, it is not clear whether (or why) the axial symmetry is *spontaneously broken*. Indeed, wrinkle domains are evident in Figs. 1b,1c, in which the confined patch is polygonal, but for a circular patch (Fig. 1d) the pattern appears similar to the axially-symmetric, ring-like wrinkles of a confined cone (Fig. 1e). The presence (or lack thereof) of spontaneous symmetry breaking may be informed by comparing ${\varepsilon_{\rm res}}^{\rm (domains)}$ found in [2] (above), to an analogous estimate of ${\varepsilon_{\rm res}}^{\rm (rings)}$, obtained by minimizing energy over axially-symmetric, ring-like deformations.

• The multi-scale nature of wrinkle patterns, where characteristic scales of wrinkles, grain boundaries, and domains, span the interval $t\leftrightarrow D$, makes a numerical study of the limit $\frac{D}{\sqrt{R\cdot t}}\to\infty$ a daunting task. One may thus hope that experiments with ultrathin polymer sheets, for which this ratio may be tuned to a few thousands or more, will overcome boundary effects and reveal the "true" ground state. However, in such conditions the surface tension, γ_{lv}, which pulls on the solid's edge, often has a non-negligible effect. Contrary to a claim made in [2] (Supplementary information), the importance of surface tension is not related to the tensile strain, $\frac{\gamma_{lv}}{Y}$, which may be tiny, but rather to the ratio $\frac{\gamma_{lv}}{\sqrt{BK}}$ (or more generally: $\frac{\gamma_{lv}}{Y}\cdot{\varepsilon_{\rm res}}^{-1}$), which is recognized as a "*softness*" parameter [4]. It is the small value of this parameter in the experiments of [2], rather than small $\frac{\gamma_{lv}}{Y}$, that underlies their observation of negligible surface tension effect. For stiff polymers (*e.g.* poysterene) with sub-micron thickness, the softness ratio may be extremely large (while $\frac{\gamma_{lv}}{Y}$ remains small), suggesting that experimenters may find a strikingly different morphology than the one reported in [2].

Finally, let's assume that rigorous energetic bounds, or careful simulations, find that the ground state, for sufficiently thin shells and away from any axial-symmetry-breaking boundaries, is not wrinkle domains, but a far simpler axisymmetric state, similarly to Fig. 1e. Will such a possible result revoke the merit of an effective smectic theory?

Quite the contrary!

To start with, the remarkable agreement between simulations and effective smectic predictions for the width and structure of domain walls [2], unequivocally demonstrates the predictive power of this approach for situations where wrinkle domains are guaranteed to exist (*e.g.* near flat boundaries). From this perspective, even though the bulk state may not consist of wrinkle domains, the effective smectic theory [2] is a valuable tool for studying meso-scale morphological features in the vicinity of boundaries. One must not forget that in such a scenario the morphological signatures at larger scales, most importantly the domain size, L_{dom}, may not be properly described by the prediction of [2] (above). Instead, the size of wrinkle domains is likely to be determined by the shape of the boundary (*e.g.* Fig. 1c).

Next, one may ask whether an extension of the effective-smectic approach may be applied for a coarse-grained description of patterns different from wrinkle domains. To wit, let us note that underlying the effective smectic theory is the assumption of a "domain director", $\hat{n}(\mathbf{x}) \approx \hat{n}_0$ ($\mathbf{x}$ in a given domain) that characterizes the compression direction in each domain. Deviations from $\hat{n}_0$ underlie the smectic energies (of bending and compression) [2]. However, inspection of Figs. 1a,1e, reveals a compression direction that varies continuously, with bend ($\hat{n}\times\nabla\times\hat{n}\neq 0$, Fig. 1a) or splay ($\nabla\cdot\hat{n}\neq 0$, Fig. 1e). A coarse graining approach, which penalizes deviations from such director fields, is likely to include *effective nematic* energies.

Another complexity, pertains to the assumption of a "slaving" condition, which invokes that the ratio, $\frac{A}{\lambda}$, between the wrinkle amplitude, $A(\mathbf{x})$, and its wavelength, λ, is completely determined by the confining geometry [2]. This echoes standard tension-field-theory, in which the stress field underlying wrinkles is assumed to be "pre-determined" by the confining geometry and an external tensile load, through minimization of a "dominant" energy that approaches a constant value (namely, $\frac{\varepsilon_{\text{res}}}{\varepsilon_{\text{bare}}}\to Cst$) as $\frac{D}{\sqrt{R\cdot t}}\to\infty$ [1, 3]. However, recent studies [6] indicate that under pure geometrically-incompatible confinement, such a slaving condition may no longer be valid, implying that a coarse-grained energy functional should include an explicit term that couples the amplitude, $A(\mathbf{x})$, and the director, $\hat{n}(\mathbf{x})$.

Those who will follow the path marked by Aharoni *et al.* [2] may thus discover a virgin territory in which new, yet unknown versions of liquid crystal theory become effective coarse-grained models for deformations of thin solids in geometrically-incompatible confinement. From this perspective alone, this paper is an asset to those who strive to see universality in the complex morphology exhibited by a candy wrapper or an inflated mylar balloon.

References

[1] J. Hure, B. Roman and J. Bico, Phys. Rev. Lett. **109**, 054302 (2012).

[2] H. Aharoni *et al.*, Nat. Comm. **8**, 15809 (2017).

[3] E. Hohlfeld and B. Davidovitch, Phys. Rev. E. **91**, 012407 (2015).

[4] J. Huang *et al.* , Phys. Rev. Lett. **105**, 038302 (2010).

[5] P.G. De Gennes and J. Prost, The Physics of Liquid crystals, 2^{nd} Ed. (1995).

[6] Y. Sun, B. Davidovitch and G.M. Grason (in preparation).