On shape and elasticity: bio-sheets, curved crystals, and odd droplets

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Summary

It is not possible to fold a regular sheet of paper in half – top to bottom and side to side – more than seven times, for it then becomes too thick, as compared to the length and the width, to halve it once more. This simple hands-on exercise illustrates the connection between the thickness of an object and the ease with which it can be bent out of plane. Indeed, thin solid systems such as the initial single-layer paper can be reshaped relatively easily, allowing the creation of all kinds of three-dimensional structures from effectively two-dimensional materials. The shape of deformable thin solids is therefore an expression of their mechanics, or in other words, a result of the external forces applied and/or internal interactions within the material.

In this thesis, we dive into physical phenomena related to shape deformations of thin sheets and hollow shells on the micrometer scale, namely: open sheets built from a molecule found in living cells, two-dimensional crystalline shells, and the interface of oddly-shaped droplets. These systems, besides being interesting on their own and also of interest in fields beyond fundamental physics, are prime instances to study the effect of geometry, global and local, in the mechanics of two-dimensional media. And vice versa, it is fun to look at what unexpected shapes arise from the interplay of a few mechanical ingredients. Being elastic solids, they offer resistance against local deformations away from a reference state, expressed as internal material stresses. Being thin, and thus easy to deform along the vanishing dimension, the cost of in- and out-of-plane deformations can be decoupled. Their elastic energy has two contributions: (i) the stretching energy cost of compressing or extending the surface, similar to a bed sheet being adjusted to a flat mattress, and (ii) the bending energy cost of reshaping out of flatness, similar to rolling a piece of paper. In reality, the elasticity of the systems studied in this work is somewhere between paper and fabric; they offer some resistance to both stretching and bending the surface when they are reshaped.

In many cases, deformations on thin systems arise as a consequence of local prestress, for instance, due to the geometrical arrangement of the material constituents or their interactions. An interesting example included in this thesis is the assembly of tubulin structures with encoded spontaneous curvature, i.e., the surface curvature that reduces the stress and minimizes the bending energy. Tubulin is a round protein present in almost all living cells, where it pairs up in tightly bound dimers. These dimers can assemble into a variety of larger structures by stacking up top to bottom, like Lego blocks, and also associating side to side, thus forming thin membranes. Although flat tubulin sheets have been observed, the most common and best-researched assemblies are long hollow cylinders called microtubules, key to a myriad of biological processes. Unlike the classical Lego block, tubulin dimers are effectively asymmetric: a side-to-side lateral wedge induces the cylindrical closing while a top-to-bottom longitudinal kink in the dimer causes outward curving stress. The flexibility and the

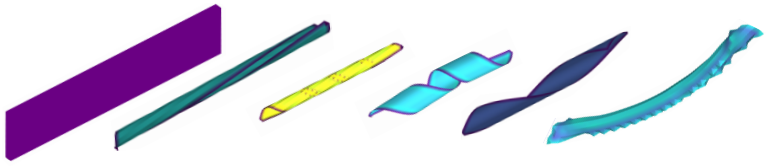

Figure S1: Bio-sheets. A thin sheet built from smaller constituents, such as biomolecules, have different geometries depending on the geometry and interactions of the building blocks. We use computer simulations to investigate these shapes.

kink in dimers are affected by the chemical environment or following our analogy, it alters the effective shape of the building blocks. As such, we think these alterations are behind the structural variability of tubulin assemblies, as several other shapes have been observed but by far less studied than microtubules. These include hoops, C-shape long sheets, multilayered rings, and helical ribbons of varying curvature. By considering a minimal model that includes the anisotropy in the intrinsic dimer curvature, we use numerical simulations to uncover the different shapes that result from changing the internal mechanics of elastic tubulin sheets (see Figure S1). This gave us insight into why cylinders and other microtubule-like shapes are prevalent in these assemblies and how polymorphism emerges.

Back to the elastic energy, although stretching and bending are decoupled in their quantification, the physical mechanisms behind them are interconnected and often in competition. For example, consider a two-dimensional crystal, that is, a solid monolayer where the building blocks are arranged in a lattice with well-defined order. In particular, think of a mesh where every point has six equally-spaced neighbors, or equivalently, a patchwork of regular hexagons. While flat sheets can have such a perfect sixfold lattice (see back cover), the same is not true for closed crystals such as a spherical shell. Because of the way a sphere curves in space, it is mathematically impossible to patch it using only regular hexagons but there are lattice points that fail to have six neighbors. This type of defect is called a disclination and its distortion causes in-plane stretching. Similar to the tubulin sheets, the closed crystal is pre-stressed, not because of the shapes of the building blocks but because of how they are arranged with respect to each other. The best way to accommodate the awkward mathematical constraint is by having twelve fivefold disclinations, as distant as possible from each other (see Figure S2). The staple illustration for geometrical frustration on a sphere is therefore the twelve pentagonal patches on a soccer ball. However, this does not fully relax the elastic stress, and other stress-screening mechanisms have been observed for spherical crystals. One of them is the outward buckling of the surface around the disclinations at the expense of increasing the bending energy. The result is the transformation of a sphere into an icosahedron – a polyhedron with 12 vertices – in order to deal with the angular gap left by a missing neighbor. If the sphere is small compared to the distance between neighboring points (low-density crystal), thus highly curved, buckling is not worth the bending cost. However, the disclination stress scales with the number of lattice points, and larger crystals then express icosahedral shapes. The shell encapsulating the genetic material of viruses is a well-studied instance of this

Figure S2: Curved crystals. A crystal with sixfold symmetry is mathematically bound to have fivefold defects referred to as disclinations (red centers), such as the twelve black pentagonal patches on a soccer ball, regardless of the number of lattice points. Even when the lattice structure cannot be resolved in much denser crystals, like the frozen interface of deforming emulsion droplets, the stress from the inevitable disclinations is felt across the surface, represented by the brighter regions. We explore two screening mechanisms, where increased curvature by buckling, here highlighted, is one of them.

mechanism: smaller viral capsids are spherical while larger ones are icosahedral. Yet, for much denser crystalline assemblies, the immediate region around the disclination looks comparatively flatter (similar to "flat" bike roads on a round Earth). In these, a second screening mechanism has been observed. Perhaps surprising at first, another way to alleviate the stress introduced by the disclinations on a sphere is to form even more defects than the minimum necessary. The trick is which and where. The distortion caused by the initial disclination can also be screened by forming lines of alternating fivefold and sevenfold defects radiating from it. These are referred to as dislocation scars.

So far, we have glanced at some of the intricacies of the connection between shape and surface elasticity. However, the elastic energy is in many systems not the only relevant contribution to the total mechanics. A wonderful example is found in particular types of emulsion droplets recently explored. These are micrometer oil droplets stabilized in an aqueous solution by surfactant molecules sitting at the interface. Under the right chemistry, that interfacial monolayer can freeze while the bulk oil and water remain liquid, forming effectively a two-dimensional crystal. In contrast to a common suspended droplet which is spherical because it is governed by surface tension, these interfacially-frozen droplets undergo a surprising set of shape transformations when the system is progressively cooled down since the surface tension then decreases. The initial round shape facets into an icosahedral one, which then flattens into a hexagonal platelet, and further deforms into other curiously specific polyhedral flat shapes (see Figure S3). In this thesis, we unravel the physical mechanisms behind the first two steps: the faceting and flattening of the droplets. Similar to deforming a water balloon, the different droplet geometries are a result of deforming the thin interface, so we build a model to describe their two-dimensional mechanics and gain further understanding of the physical phenomena.

Although the initial droplet faceting to an icosahedral shape places the disclination-buckling mechanism as a prime suspect, there were two subtleties to be addressed. The first one is related to the very dense crystal formed by the frozen interface. Because of the size ratio between the droplets and the surfactant molecules, dislocation scars are expected. Yet, buckling was observed and unfortunately the actual lattice is way too small to be resolved with microscopy. Furthermore, the theoretical

or computational models available to study defect stress on spherical crystals were based on systems with considerably fewer lattice points and thus unfeasible in our study. This motivated setting up a new framework that accounts for the connection between curvature and stress relief around a set of discrete disclinations which are nonetheless surrounded by an arbitrary but large number of other dislocation scars. The second subtlety is that, unlike viral capsids, small droplets are icosahedral while large ones remain spherical. However, having established a framework appropriate for the dense crystalline interface and after a closer look at the dependence of the deformation pathway with the droplet size, our model unraveled there are in fact four key ingredients at play: (i) Temperature-modulated surface tension, (ii) buoyancy pressing the droplets against a top cover of the microscope slip, and the elastic component from the solid interface including (iii) spontaneous curvature of cone-shaped surfactant molecules and (iv) in-plane stress of the inevitable defects on the spherical crystal; the whole package.

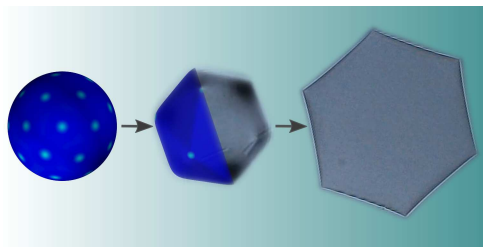

Figure S3: Odd droplets. Micrometer liquid droplets undergo fascinating shape transitions when stabilized by an interfacial frozen monolayer, including icosahedral shapes and flat hexagonal platelets. We study the mechanics of thin membranes, such as their solid interface, using computational three-dimensional modeling.

The monolayered closed crystals, the frozen interface of emulsion droplets, and the polymorphic tubulin sheets are all, at heart, fascinating expressions of the interplay between geometry and mechanics in elastic thin solids. Therefore, in **Chapter 1** we start with a conceptual introduction to the description of the shape and the elasticity of solid membranes, and to how they are intimately linked through the microscopic details of the material constituents. In **Chapter 2**, we give formal mathematical definitions to these concepts for two-dimensional surfaces in three-dimensional space, which are the main study subjects in this thesis. On a basis of differential geometry and linear elasticity theory, we build the models behind the stretching and bending energies of curved surfaces, and we outline how these translate to discretized surfaces, needed for computational 3D modeling. In **Chapter 3**, we build and explore the semi-continuous model of defects of dense crystals on closed geometries, including icosahedral shapes. Besides generalizing previous results obtained for spherical crystals, the model implementation could further be used in the study of oddly-shaped droplets. In **Chapter 4**, we outline our simple but rich study on the mechanics of deforming emulsion droplets and we explain the insightful results in the context of previous work done. Last but not least, **Chapter 5** presents the results on the expected and unexpected shapes a bio-sheet can obtain under varying internal stress expressed as spontaneous curvature, the first computational study on tubulin polymorphism.