Edge Effects Determine the Direction of Bilayer Bending

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Supporting Information

ABSTRACT: We elucidate the reason for preferential bending along the long edge in thin rectangular bilayers in which one of the layers is isotropically strained. While this preference has been observed previously, the physical basis for this preference has not been understood. We find that the bending direction is determined by the existence of doubly curved regions at the curled edges, which lower the energy. This energy difference between “spiral” and “cigar” shapes increases with aspect ratio.

KEYWORDS: Bilayer, edge effects, bifurcation, doubly curved, self-folding, polypyrrole

There has been considerable recent interest in understanding spontaneous curvature, such as seen in the deformation of bilayers.1–3 Bilayers are planar membranes or plates with a different material on each face, and they bend or wrinkle into curved three-dimensional structures if one face expands more than the other. Similarly, a single material (unimorph) will bend in response to a strain gradient in the thickness direction. One of the motivations for studying curvature arises because combination of top-down patterning via photolithography with bottom-up design of material structures capable of spontaneous or induced curvature allows the fabrication of structures that cannot readily be created by conventional microfabrication,4 and it is envisioned that these structures will allow new functionalities. Another motivation is that this bending phenomenon is seen on every length scale, from macroscopic bilayer thermostats to nanoscale graphene.5

Microscale bilayers have been constructed from a wide variety of organic and inorganic materials.5 Recent examples are light-actuated microrobots built from rectangular bilayers that include a liquid crystalline polymer,6 silicon microelectromechanical systems (MEMS) that fold into three-dimensional shapes under thermal actuation,7 magnetostrictive—piezoelectric bilayers that undergo giant magnetoelactromotricity for memories and energy conversion,8,9 and piezoelectric MEMS radio frequency switches for wireless communication.10 Cho et al. created complex structures with annular bilayers, the shapes of which depended on the annulus width;11 wrinkled, saddled, and wedge-shaped structures were formed. Often, intrinsic stress is exploited to self-assemble curled structures. For example, spirals have been microfabricated that curl upon release from the substrate due to lattice mismatch strain.11 Chun et al. formed micro- and nanoscale tubes with controlled sizes by the self-rolling of epitaxially mismatched rectangular bilayers, demonstrating that the bending direction varied depending on the length to width ratio (the aspect ratio A).1 They also performed FEM modeling, showing that the strain energy was lower for long-side rolling than short-side rolling, but the reason for the lower energy was unclear. Cendula et al. looked at the rolling of wrinkled, strained rectangular films, finding a preferential rolling direction that depended on the geometry and strain gradient.3 Despite the extensive prior work, however, a fundamental elucidation of the mechanism that propels rolling in the long direction is still missing.

For a bilayer to become curved in two orthogonal directions, like the surface of a sphere, it must undergo stretching, which for large deformations has a significant associated stretching energy.12,13 So, the plate instead tends to assume shapes that are locally curved in one direction only (such as cylinders and cones), minimizing regions of double curvature. A few of these bending possibilities are illustrated in Figure 1A based on experimental observations; other configurations are also possible.1–3 The plate could bend into a “cigar” (short edge curved), a spiral (long edge curved), or a corkscrew14 (intermediate angle). These configurations have the same curvature in the direction of bending, so one might expect them to have the same elastic energy. If this were the case, the bending direction would be random, which is not observed experimentally. (The radius of curvature has been shown to depend on the strain, thickness, and moduli of the layers, and not the bilayer dimensions.1) Alternatively, the plate can develop “dog-ears”, with the corners curved inward to the point at which they meet (see for example the Supporting Information).

In this paper, we offer new insight into why a rectangular bilayer will curve along the long direction. We show mathematically that the reason lies in the spatial distribution of curvatures. The bilayer does not bend into a perfect tube, but has narrow regions of double-curvature at the curved edges that lower the energy density at those locations. The total length of curled edge is greater for bending in the long direction, so during the transition from a spherical to a
singly curved shape, it is more favorable for the rectangle to adopt the spiral geometry. The edge effects become relatively stronger as the length to width ratio increases. The increasing probability of curvature along the long direction with aspect ratio \( A \) is confirmed experimentally using isotropic bilayers of polypyrrole and gold, and the slight double curvature is also shown.

Hou and Chen studied, experimentally, rectangular bilayers that were clamped along one edge, which constrained the direction of bending. They found that at intermediate aspect ratios, much of the bilayer was doubly curved, whereas at large and small ratios it was nearly singly curved except for a narrow region near the edges. (We have also simulated this problem and obtained results that are essentially identical to theirs, as shown in the Supporting Information.) In this work we consider bilayers that are 2 orders of magnitude thinner and without the clamp at the edge, removing the bias on the bending direction.

The classical model of bilayer bending, initiated by Timoshenko in 1925 and since extended, assumes that the bilayer can bend in only one direction and results in a bilayer with uniform curvature. More recent models have considered bilayer bending in two dimensions. Mansfield found analytical solutions for large deflections of circular and elliptical plates having lenticular cross sections with a temperature gradient through the thickness. For small gradients, the plates formed spherical caps, curved equally in all directions. At a critical gradient, a configuration with greater curvature in one direction became more favorable. Because of the lens-shaped thickness profile, even though the elliptical plate had a major axis it showed no preferred direction for bending even for large deflections. For the uniform thickness typical in experiments, we show that a particular bending direction is preferred. For a circular bilayer of uniform thickness, Freund determined the strain at which the spherical cap becomes unstable using low order polynomial solutions and finite element simulations. Our model extends that of Freund to more general bilayer displacements and arbitrary values of layer thicknesses.

We model the bilayer as two rectangular layers of elastic material: a passive substrate and an active film (Figure 1B). Each has a length \( L \) in the \( y \) direction, width \( W \) in the \( x \) direction, Young’s moduli \( E_i(E_f) \), thicknesses \( h_i(h_f) \), and Poisson ratio \( \nu \), and they are bonded together at \( z = 0 \). In the model, an isotropic actuation strain \( \varepsilon_a \) is applied to the film; this is the strain that the film would experience if it were free, and a simple expansion (or contraction) by \( \varepsilon_a \) in \( x, y, \) and \( z \) would result in the film having the lowest energy. However, the film is bonded to the substrate, which is not actuated, and the two layers must have the same deformation, \( u_i \) at their interface. The interfacial strain that minimizes the total elastic energy of the bilayer therefore lies somewhere between \( \varepsilon_a \) and 0. The total energy can be decreased further if the bilayer bends into the \( z \) direction because in a curved plate, the layer at the outer circumference is stretched while the one at the inner circumference is compressed, bringing each layer closer to its individual “preferred” strain.

Our model considers the elastic energy of a bilayer as two plates undergoing moderate out-of-plane deflections, intermediate between the thickness and the width, with continuous deformations at their interface (\( z = 0 \)). This is well described by the Föppl–von Kármán equations:

\[
U = \int_{-1/2}^{1/2} \int_{-1/2}^{1/2} \int_{-h_i}^{h_i} \frac{E(x,y,z)}{2(1+\nu)} \left( \frac{1}{1-2\nu} \nabla^2 u_i(x,y,z) + \frac{1}{1-2\nu} \nabla^2 u_f(x,y,z) \right) \, dy \, dx \, dz
\]

(1)

Here \( \nabla^2 \) is the Laplacian in the \( x \) and \( y \) directions, \( \nabla^2 \) is the shear strain. The strains are given in terms of the displacements \( u_i, u_f, u_z \) of the bilayer in the \( x, y, \) and \( z \) directions.

\[
\nabla^2 u_i(x,y,z) = -z \frac{\partial^2 u_i}{\partial x^2} (x,y) + \frac{1}{2} \left( \frac{\partial u_i}{\partial x} (x,y) + \frac{\partial u_i}{\partial x} (x,y) \right)
\]

(2)

In (2), \( x_i \) and \( x_f \) range over \( (x_i, y) \) while \( u_i \) and \( u_f \) range over \( (u_i, u_f) \), respectively. The first term on the right side of (2) is due to bending and the second is due to in-plane stretching. The third is the leading nonlinear stretching term that occurs for small-to-moderate strains (although the materials obey linear elasticity, strain depends nonlinearly on displacement; see ref 23). The fourth term is the equilibrium strain \( \varepsilon (z) \), which equals \( \varepsilon_a \) in the film and zero in the substrate. (1) and (2) are well-known equations of elasticity, and versions of these equations have been used recently to study single thin plates under forcing.12,13,24

For a given \( \varepsilon_{aw} \), we obtain the bilayer shape by finding the displacements that minimize the elastic energy in (1). We discretize the integrals over \( x \) and \( y \) using uniform grids, and we compute the \( z \) integral analytically. We use Newton’s method to find the energy-minimizing shapes, using analytical formulas for the gradients and Hessian matrices. (This approach allows a more direct knowledge and verification of the numerical accuracy and convergence rates of our results than would be obtained using commercial software.) To compare with our experiments,
Figure 2. (A) From the simulations, curvature as a function of actuation strain \( \varepsilon_a \). The curvatures in the \( x \) (blue) and \( y \) (orange) directions, averaged over the bilayer, are plotted for \( A = 1.001, 1.01, 1.1, 2, \) and 10. The insets show shapes for \( A = 2 \) at the indicated \( \varepsilon_a \), the out-of-plane \( z \) axis is greatly exaggerated for illustration. The colors go from blue to red as the \( x \) displacement increases. For comparison, an alternative equilibrium state with higher energy, the cigar (which does not correspond to any of the lines of the plot), is shown at the same \( \varepsilon_a \) (0.02\%) as the adjacent spiral. (B) Maps of curvature \( \kappa \) in \( x \) and \( y \) directions for three \( \varepsilon_a \), spanning the transition from spherical cap to developable spiral for \( A = 2 \). The colors go from blue to red as the curvature increases. (C) Maps of elastic energy per unit area for the spiral and cigar at \( \varepsilon_a = 0.1\% \), as a fraction of the energy density for the Timoshenko solution. (D) Total energy difference \( U_{\text{spiral}} - U_{\text{cigar}} \) vs \( \varepsilon_a \) for different \( A \).
nearly equal, giving a nearly spherical cap. Quadrupling $\varepsilon_x$, $\kappa_y$ becomes large and nearly uniform while $\kappa_x$ drops dramatically, producing a spiral, modified by regions of double curvature near the longer sides (the edges bend slightly inward). Quadrupling $\varepsilon_y$ again, the regions of nonzero $\kappa_x$ become narrower. These regions are similar to other edge layers in solid mechanics, which can occur when a control parameter (here, $\varepsilon_y$) becomes sufficiently large. Previous bilayer models had insufficient resolution to show the edge layers. However, the experiments of Hou clearly showed their presence for small $A$. Similar edge layers occur for the cigar, except that they lie on the shorter sides. These edge effects become relatively stronger as the length to width ratio increases.

The effect of edge layers on the elastic energy component is shown in Figure 2C. The color maps give the local elastic energy per unit area, as a percentage of the Timoshenko solution energy density, which was used as a baseline. Away from the edge layers, where the bilayer is curved primarily in one direction, the energy density is within 2.5% of that of the Timoshenko solution, but in the edge layers the energy density is nearly 20% lower. Because the edge layers occur on only two of the four sides, and because they are longer for the spiral than for the cigar, the energy of the spiral is lower.

In Figure 2D we plot the difference in total energy between the spiral and the cigar, relative to that of the Timoshenko solution. The energy difference is largest at $\varepsilon_y$ just beyond the transition from spherical cap to developable shape. At larger $\varepsilon_y$, the edge layers become smaller and contribute proportionately less to the total energy. The energy difference is larger for larger aspect ratios but is still fairly small (<1.6%) at $A = 10$.

Because the elastic energy difference between the spiral and cigar configurations are minute, to study the bending question experimentally requires bilayers that are essentially perfectly homogeneous in the plane of the layer, so that no directional bias is introduced that could influence the bending direction. Macroscale fabrication methods such as roll milling to produce sheet metal or extrusion to produce polymer films introduce a definite anisotropy to the material. For this reason, and because of the significant role of bilayers in micro- and nanosystems, bilayers were produced using surface microfabrication techniques (Figure 3A). To achieve isotropic mechanical behavior, the films of the bilayer were amorphous or polycrystalline: the active film was polypyrrole (PPy), and the substrate layer was gold (Au).4,27,28

PPy is an electroactive polymer that changes volume upon the application of an electrochemical potential, which allows electrical control over the strain. The Au layer served as the constant-volume substrate, as well as the electrode contacting the PPy. The PPy was doped with dodecylbenzenesulphonate (DBS) during electrochemical deposition. The bilayers were actuated in an aqueous solution of NaDBS using a potentiostat, contracting upon oxidation and expanding upon reduction. The actuation strain in PPy depends on its oxidation level via the incorporation of charge-compensating ions, which is determined by the voltage applied to the Au film (working electrode) in intimate contact with the PPy. (The rate of oxidation or reduction depends on the rate of arrival of charge-compensating ions, which may be higher at the edges.) The in-plane strain in fully actuated PPy(DBS) is on the order of 3%.28

Another critical factor was ensuring that the bilayers were able to move freely upon first actuation. It has been shown that if bilayers curl during release from the substrate by undercutting a sacrificial layer, the final rolled state is influenced by that history of constrained rolling. Constraints also affect the bending behavior of the PPy bilayers (see Supporting Information). Because the as-deposited Au and PPy films were unstressed, these bilayers remained flat during undercutting and did not curl until they were actuated, removing constraints and any memory effects.

A series of devices was fabricated (see the Supporting Information for details) having widths of 100 $\mu$m and increasing lengths to give aspect ratios between 1 and 3. To apply the electrochemical potentials, the bilayers required an attachment point to the substrate, which was a small square 16 $\mu$m wide at the center of the bilayer (Figure 3A). There were 100 bilayers of each aspect ratio in a $10 \times 10$ array. As mentioned previously, in this work the layers were unstrained during fabrication, and so remained completely flat during undercutting to remove the sacrificial layer. This is unlike in some prior work in which the membranes began curling during the etching process, which affected the final curled shapes.

Upon electrochemical actuation, the bilayers curved upward around the center mount point, as shown in the scanning electron microscope image (SEM) in Figure 3B. Between these two curved regions, on either side of the center mount, the bilayers remained flat. Panels B and C of Figure 3 show double curvature at the corners, and panels B–E of Figure 3 show double curvature at the long edges (marked by arrows), as predicted (Figure 2A). Figure 3C also shows that the bilayers bent with a uniform curvature into a circle, unaffected by the center mount and showing both that the working electrode provided the same potential to the entire device, resulting in an unvarying oxidation level in the PPy film, and that the fabrication process produced devices with the correct behavior. In Figure 3D, both sides of the bilayers curled more than 90°, giving a double-roll configuration because of the center mounting.

Figure 3. (A) Cross-sectional schematic (not to scale) of one of the bilayer actuators. The upper SiO$_2$ (gray) is a sacrificial layer that is removed in the final fabrication step to free the bilayer to bend. (B–E) SEM images of bilayers after actuation. Arrows indicate regions of double curvature at the long edges.
Figure 4. The fraction of defect-free PPy/Au bilayers that curved in the y direction (i.e., spirals for \( A > 1 \)) as a function of aspect ratio. The insets show close-up images of parts of two arrays in the fully oxidized (bent) state, an optical micrograph for \( A = 1.0 \) and an SEM for \( A = 3.0 \).

The bending direction as a function of aspect ratio is shown in Figure 4. Each point corresponds to one array (100 devices) on the sample and shows the fraction of devices in the array that bent in the y direction, which for \( A > 1 \) means that the longer edge is curved. In the array with \( A = 3.0 \) (see inset), there is a predominance of spirals. For the shorter bilayers with \( A = 1.0 \), there is a mixture of bending in the x and y directions. The sides of the shorter bilayers were curled \( \sim 90^\circ \), so they appear dark in the optical micrograph.

There are several things to note in Figure 4. First, the preferred configuration at high aspect ratios was the spiral (i.e., curved along the longer edges). This was expected from the prior experimental and theoretical work discussed above. Second, the aspect ratio had a powerful influence on the fraction of bilayers in the array that bent into a spiral versus a cigar (i.e., curved along the shorter edges). A significant increase in the fraction of y-directional bending was seen even for the smallest tested asymmetry, going from \( A = 1.00 \) to \( A = 1.01 \). (The energy difference at \( A = 1.01 \) is less than 0.03%, Figure 2D.) By \( A = 1.1 \) (at which the energy difference is still only 0.2%), over 90% of the devices were spirals. Conversely, for \( A = 1 \) bending along either the x or y directions was random. (There was actually a small bias toward curvature in the x direction in this particular A = 1 array. Other A = 1 arrays showed either no bias, a small bias in x, or a small bias in y; see the Supporting Information.)

It is also important to note that almost no bilayers bent into corkscrews or dog-ears. (This is not due to the shape of the center mount. The Supporting Information shows that bilayers with round and square center mounts had the same behavior. The Supporting Information also shows that dog-ears form when the bilayers initially adhere slightly to the surface, rather than being completely free.) In samples with a greater number of fabrication nonuniformities and defects, there were tilted corkscrews, dog-ears, and dog-ears, but with increasing A the spiral still became heavily favored, with the frequency of the other configurations dropping sharply. This finding confirms the model result that there are no stable off-axis configurations.

In conclusion, the model and the experiments found only two stable equilibria: the spiral and the cigar. The spiral was energetically preferred, and the preference for the spiral increased with aspect ratio. The model shows that edge layers are the important feature that differentiates the spiral and cigar from the constant curvature Timoshenko approximation, and which therefore differentiates their elastic energies. This double curvature at the long edges, observed in the devices, has only rarely been seen because it is a very small effect, and its energy-lowering effect has not previously been known to be responsible for the long direction bending bifurcation. Although we have focused on rectangular bilayers, edge layers are a general phenomenon occurring in other shapes,26,30 and we expect that they will also contribute to the direction of bending for other shapes. Therefore, this approach provides a fundamental basis for thinking about these problems in general.

**ASSOCIATED CONTENT**

Supporting Information. Full details on the mathematical model and the fabrication process, data from two additional samples, additional data on \( A = 1 \) arrays, additional images of the PPy/Au bilayers, images of devices that were made with a different fabrication process that left the bilayers sticking tightly to the substrate, which resulted in dog-ears, comparison of the bending behavior of bilayers with round versus square center mounts, data showing that a larger aspect ratio is needed to ensure the spiral geometry in less perfectly fabricated bilayers, and a video of the actuators. This material is available free of charge via the Internet at http://pubs.acs.org.

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