Control of Rolling Direction for Released Strained Wrinkled Nanomembrane

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Abstract: Strained wrinkled and flat nanomembranes have different bending properties when they are released from the underlying substrate. This is caused by increased bending rigidity of the wrinkled film in one direction. We provide theoretical and numerical analysis of the directional rolling of wrinkled films, which is important for positioning rolled-up tubes on the short mesa edge during fabrication.

Keywords: rolled-up, tube, wrinkle, nanomembrane, strain.

1. Introduction

Wrinkled (corrugated) paper in a cardboard structure is a famous example of using corrugations to increase bending resistance of product packages to mechanical damage. Transferring this knowledge to nanotechnology leads to practical advantages for fabrication of nanostructures. In this work, we apply a similar principle based on wrinkling of nanomembranes to control the rolling process of rolled-up micro- or nanotubes.

Increasing interest in applications of rolled-up nanomembranes or films [1-4] requires precise techniques to position them on a substrate surface. Usually, pre-strained rectangular flat films on lithographically defined mesas are partially released by etching the underneath sacrificial layer from all sides. A large strain gradient causes the films to bend and roll up into tubular structures. Usually, the rolled-up tubes form along the long edges only, see Fig. 1(a). In this work, we show that releasing an initially wrinkled film on a mesa can suppress the formation of tubes at the long (wrinkled) edges and instead cause the roll-up of the film along the short (flat) edges, Fig. 1(b).

2. Theory

We consider an elastic strained film of thickness t and dimensions L_x and L_y, where $L_y < L_x$. Linear isotropic elastic properties with Young's modulus E and Poisson's ratio ν are assumed for this film. The cartesian coordinate system (x, y, z) is defined with the origin at the middle plane of flat (wrinkled) film and with the orientation of axes given in Fig. 1(a). In the following, we show the analytical expressions for elastic and bending properties of the considered flat and wrinkled film.

2.1 Flat film

First, we consider a flat film, which is initially biaxially strained with

(1) $$\varepsilon_{xx}^{(i)}(x,y,z) = \varepsilon_{yy}^{(i)}(x,y,z) = \varepsilon_0(z),$$

(2) $$\varepsilon_{zz}^{(i)}(x,y,z) = -\frac{2\nu}{1-\nu}\varepsilon_0(z),$$

where $\varepsilon_0(z) = \bar{\varepsilon} + (\Delta\varepsilon / t)z$ and $\bar{\varepsilon}, \Delta\varepsilon$ are average strain and strain gradient in the film, respectively. Since the film is thin, normal z-stress through the thickness is zero and eq. (2)

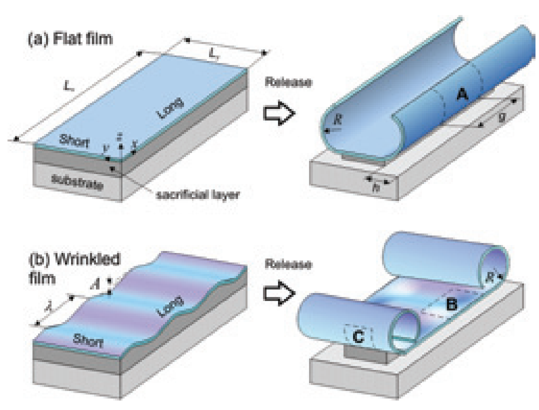

Figure 1. (a) Initially, a flat film rolls up into tubes at the long edges after partial release from the substrate. In contrast, (b) a wrinkled film relaxes into tubes at short edges after partial release. Actual film portions A, B, and C taken for our simulations are denoted as dashed regions and they have the same dimensions (h×g).

follows from Hooke's law. All shear strains are zero. The initial strain considered here can be caused by epitaxial mismatch strain, different thermal expansion of substrate material and the film, and other thin film fabrication conditions. In experiment, when the sacrificial layer is etched away below the film from all sides, the strained film becomes free-standing and it can relax the initial strain by bending into tubular structure, see Fig. 1(a), with radius of curvature for bending in both *x* and *y* directions given by [5]

$$R = \frac{D}{M} = \frac{t}{(1+\nu)\Delta\varepsilon}, \tag{3}$$

where $D = Et^3/(12(1-\nu^2))$ is the flexural rigidity of the film and $M = E\Delta\varepsilon t^2/(12(1-\nu))$ is the bending moment per unit length caused by the strain gradient through the film thickness. This result can be alternatively obtained also by minimizing the elastic energy of the film with tubular shape assumption [4]. In the experiment, the tube actually forms on the long edge only, because rolling up of the film on the long edge relaxes more elastic energy of the initial strained film.

2.2. Wrinkled film

For the wrinkled film, we assumed the same elastic constants of this film as the flat film considered in the former subsection, Fig. 1(b). The wrinkle shape is given by deflection along *x*-direction: $\zeta(x,y) = A\sin(2\pi x/\lambda)$, where A and λ are the amplitude of the wrinkle and its wavelength. The initials strain are given by

$$\varepsilon_{xx}^{(i)}(x,y,z) = \varepsilon_{yy}^{(i)}(x,y,z) = \varepsilon^{w}{}_{0}, \tag{4}$$

$$\varepsilon_{zz}^{(i)}(x,y,z) = -\frac{2\nu}{1-\nu}\varepsilon^{w}{}_{0}, \tag{5}$$

for $\varepsilon^{w}{}_{0}(x,y,z) = \bar{\varepsilon} + (\Delta\varepsilon/t)(z-\zeta(x,y))$ given linearly from the middle plane of the wrinkled film. The wrinkled (corrugated) film has obviously different elastic and bending properties in two perpendicular directions (*x* and *y* directions). Therefore, classical work on anisotropic plates [6-7] uses an equivalent orthotropic flat plate to account for different elastic and bending properties of wrinkled film in these 2 directions. The Hooke's law for orthotropic plate is written as

$$\begin{pmatrix}\sigma_{xx}\\ \sigma_{yy}\end{pmatrix} = \frac{1}{1-\nu_{xy}\nu_{yx}}\begin{pmatrix}E_x & \nu_{xy}E_y\\ \nu_{yx}E_x & E_y\end{pmatrix}\begin{pmatrix}\varepsilon_{xx}\\ \varepsilon_{yy}\end{pmatrix}. \tag{6}$$

For sinusoidal wrinkles with small aspect ratio $A \ll \lambda$, the expressions for Young's moduli E_x, E_y and Poisson ratios ν_{xy}, ν_{yx} are [8]

$$\begin{aligned} E_x &= \frac{E}{1+6(1-\nu^2)A^2/t^2}, \quad E_y = E, \\ \nu_{xy} &= \frac{\nu}{1+6(1-\nu^2)A^2/t^2}, \quad \nu_{yx} = \nu. \end{aligned} \tag{7}$$

The wrinkled film is softer in *x* direction than the flat film, as intuitively expected. For bending (flexural) rigidity, we have

$$\begin{aligned} D_x &= D, \\ D_y &= D\left(1+6(1-\nu^2)A^2/t^2\right). \end{aligned} \tag{8}$$

The effect of enlarged bending rigidity on bending radius leads to

$$\begin{aligned} R_x &= R, \\ R_y &= \frac{D_y}{M_y} = R\left(1+6(1-\nu^2)A^2/t^2\right) > R, \end{aligned} \tag{9}$$

where $M_y = E_y\Delta\varepsilon t^2/(12(1-\nu_{yx}))$ is the bending moment in *y* direction [9]. The bending radius for bending from long edge (in *y* direction) is also larger than for bending from short edge (in *x* direction). For comparison with the result obtained from the finite element simulations, we compared the film curvature along *y*-direction $\kappa_y = 1/R_y$.

3. Method

For simplicity, we compare only the elastic relaxation of released flat and wrinkled film stripe portions as indicated by the dashed regions in Fig. 1. One edge (long or short) is fixed to the sacrificial layer, lateral edges of stripes A and B are connected by periodic boundary condition for displacement, and all other boundaries are free. We use COMSOL Multiphysics version 3.5a with Structural Mechanics Module (solid, stress-strain) in three dimensions with large deformation for this work, where the Green's strain is given by [10]

$$\varepsilon_{\alpha\beta} = \varepsilon_{\alpha\beta}^{(i)} + \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial \beta} + \frac{\partial u_\beta}{\partial \alpha}\right) + \frac{1}{2}\frac{\partial u_\gamma}{\partial \alpha}\frac{\partial u_\gamma}{\partial \beta}, \tag{10}$$

where u_α is the displacement of the point in the film and $\alpha, \beta, \gamma = x, y, z$. The first term is initial strain, the second term is a usual linear strain and the last term is caused by large deformations. Solution of equations of mechanical equilibrium gives the shape of film which best relaxes the initial strains. Large initial strains ($\approx 10^{-3}$) and large deformations make the problem highly nonlinear and we have to solve it with parametric solver by incremental increase of initial strains in small steps. The solution for smaller parameter value is used as initial guess for next parameter step to ease the solution convergence.

The geometry of flat film model is set up with initial strains described in section 2.1. The simulation of the wrinkled film relaxation consists of two parts. First, the flat film is deformed to the wrinkled state by imposing a sinusoidal displacement of amplitude A and wavelength λ on the bottom face and attaching the Deformed Mesh (ALE) Module to the film domain. Second, the wrinkled geometry iscreated from the deformed mesh and the model is set up with initial strains from section 2.2 (Eqs. (4) and (5)).

4. Simulation parameters

The film represents an experimental Molybdenum sample with a thickness of $t = 50$ nm, Young's modulus of $E = 330$ GPa and Poisson's ratio $\nu = 0.3$. The strain gradient is extracted approximately from scanning electron microscope (SEM) images, where the radius of tubes was measured as $R = 8\ \mu$m. According to eq. (3) this gives $\Delta\varepsilon = 0.5\%$. We take the average strain $\bar{\varepsilon} = -0.25\%$ to give zero initial at the middle plane of the film. The dimension of simulated stripe portion is given by etching depth $h = 6\ \mu$m to give approximately 1/8 of tube full rotation and by one period of the wrinkled film, $g = \lambda = 10\ \mu$m.

Since the film is very thin ($t/g = 0.005$), we could not use the default free tetragonal mesh, but we had to scale it in z direction to obtain reasonable number of mesh elements. For the presented results, we used 'Coarser' free mesh with scaling by 2 in z. The number of tetrahedral elements is ~3000, minimum element quality is ~0.09 and the number of degrees of freedom is ~56000.

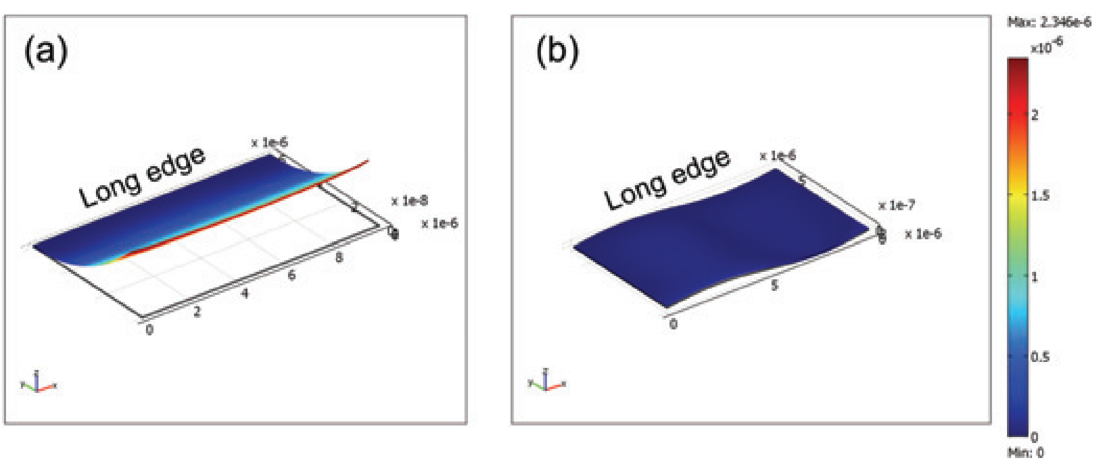

Figure 2. Relaxation of film portion fixed on long edge. (a) Initially flat film forms a tubular structure, whereas (b) initially wrinkled film ($A = 200$ nm) shows no signs of tube structure.

5. Results

We study the deformation of the flat and wrinkled film portions towards bending and tube formation. The portion of the flat film at the long edge (denoted A in Fig. 1) forms into a tubular structure with an average radius $R = 8.0$ µm (see Fig. 2(a)), which agrees well with the experimental value mentioned above. In contrast, the wrinkled film portion (denoted B in Fig. 1) deforms into a wrinkled configuration similar to the initial wrinkled film (see Fig. 2(b)), refusing to form a tube at the long edge.

For comparison with eq. (9), we extract the average curvature of radius in *y* direction κ_y over the film area as function of wrinkle amplitude *A* on Fig. 3. For flat film ($A = 0$ nm), both values overlap giving the reciprocal value of $\kappa_y = 1/R \approx 0.13\ \mu\text{m}^{-1}$. Qualitatively, curvature κ_y from the finite element calculations and theory both rapidly decrease for increasing *A*, which means that the stripe stays nearly flat and does not bend to tubular structure. For the presented geometry and parameters, we have found that the minimum wrinkle amplitude needed to suppress the bending to tube structure is as little as 3 nm.

By releasing the wrinkled film at the short edge of mesa as shown in Fig. 4 (denoted as C in Fig. 1), the tube can form conveniently as in the case of flat film with radius $R = 8.0$ µm. There is no hindrance of bending on this edge, because the structure of both the bent tube and the original wrinkle is uniform in *y* direction.

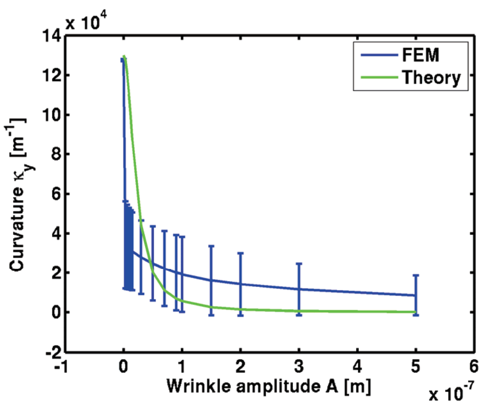

Figure 3. Comparison of stripe average curvature κ_y for rolling from wrinkled edge, extracted from finite element simulation (blue) and predicted by classical theory of eq. (9), green. Values from the simulations are shown with error bars giving standard deviation of curvature over the film area.

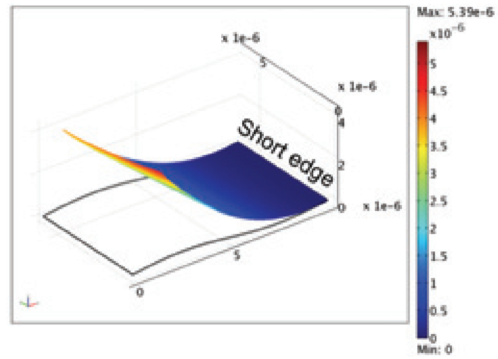

Figure 4. Relaxation of wrinkled film ($A = 200$ nm) fixed on the short edge leads to rolled-up tube structure.

As a result, when the wrinkled film is released from all four sides simultaneously, as schematically shown in Fig. 1(b), the tubes form at the short edges since bending into tubes is suppressed at the long edge.

7. Conclusions

We have shown that by introducing wrinkles to the initially strained film, the rolling direction can be controlled. Typical rolling along the long edge of the mesa can be suppressed by the wrinkle structure. Therefore, predefinition of the rolled-up tube position prior to the release of strained layer is feasible by this concept. We believe that the experimental realization of this idea is feasible in the near future.

8. References

[1] O. G. Schmidt and K. Eberl, Nature (London), **410**, 168 (2001).

[2] L. Xiuling, J. Phys. D: Appl. Phys., **41**, 193001 (2008).

[3] Y. Mei et al., Adv. Mater., **20**, 4085 (2008).

[4] P. Cendula, S. Kiravittaya, Y. F. Mei, Ch. Deneke, and O. G. Schmidt, Phys. Rev. B, **79**, 085429 (2009).

[5] Y. C. Tsui and T. W. Clyne, Thin Solid Films, **306**, 23 (1997).

[6] S. Timoshenko and S. Woinowsky-Krieger, Theory of Plates and Shells, p. 367, McGraw-Hill, New York (1987).
[7] S. Lekhnitskii, Anisotropic plates, p. 278, Gordon and Breach, New York (1987).
[8] L. X. Peng et al., Int. J. Mech. Sci., **49,** 364 (2007).
[9] Derivation will be given elsewhere.
[10] COMSOL 3.5a, Structural Mechanics Module User's guide.

9. Acknowledgements

We are thankful for discussions with J. I. Mönch.