

Supplemental Information for “Liquid dewetting under a thin elastic film”

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S1 Straining of elastic films

The home made straining apparatus is depicted in Fig. S1(a). The apparatus consists of a 250 μm thick Elastosil sheet which has been cut into a rounded plus sign shape, but contains a circular hole at its center. The Elastosil film is clamped at its four sides and supported by four posts which are able to translate in the directions indicated by arrows in Fig. S1(a). The Elastollan sample is placed atop of the hole in the Elastosil, which immediately causes the the Elastollan film to form strong contact with the Elastosil, allowing the mica to be peeled off and removed. Thus, a thin Elastollan film is left free-standing over the hole in the Elastosil.

Upon transfer, the Elastollan films have a small ($< 5\%$) pre-strain due to being stretched by the surface tension of the water during the sample preparation. In order for the initial condition to be an unstrained film, we must relieve this pre-strain. This is done by slowly bringing all four supports inwards to shrink the size of the Elastosil hole. At the point that the pre-strain is relieved, wrinkles just begin to appear in the Elastollan film. This point serves as the initial condition for the Elastollan film. To then strain the Elastollan film, the four posts supporting the Elastosil are moved outwards to stretch the Elastosil film, and hence, expand the hole at its center. To generate isotropic tension, each support is moved an equal distance such that the Elastosil hole remains circular in shape (Fig. S1(b)). For the biaxial samples, one set of supports is held fixed, while the other set of supports is displaced in the orthogonal direction. This generates a hole in the Elastosil with an elliptical shape (Fig. S1(c)). For the case of isotropic tension, the strain is found by $\epsilon = (d_f - d_i)/d_i$, where d_i and d_f are the initial (i.e. the state after the pre-strain has been relieved) and final diameters of the free-standing Elastollan film. In the case of biaxial tension, the strain along the high-tension direction is evaluated as $\epsilon_{\text{high}} = (d_{\text{high},f} - d_i)/d_i$, where $d_{\text{high},f}$ is the final diameter of the free-standing Elastollan film along the high-tension axis.

Next, the PS sample is brought into contact with the strained Elastollan film. The Elastollan adheres strongly to the PS and also to the bare silicon frame where the PS has been removed. Using a scalpel, the excess Elastollan is cut to free the sample from the straining set up. At this point, the sample looks as depicted in Fig. S2.

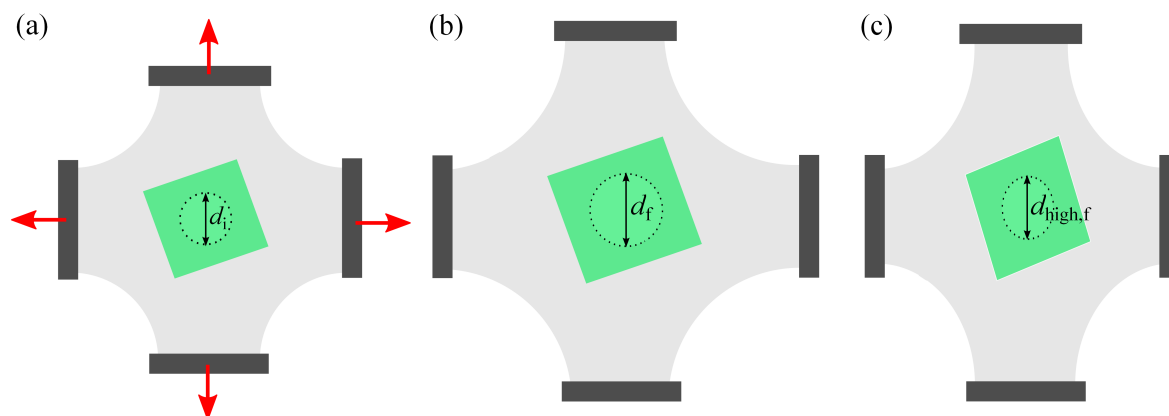


Fig. S1 (a) Top view schematic of the straining apparatus. The Elastollan film is free-standing over the hole in the Elastosil with diameter d_i . The clamped sides of the Elastosil can be translated along the axes indicated by the arrows. (b) An Elastollan film which has been strained equally in both directions (isotropic tension) to a final diameter of d_f . (c) An Elastollan film which has been strained biaxially to a final diameter $d_{\text{high},f}$ in one direction, while being held fixed in the orthogonal direction.

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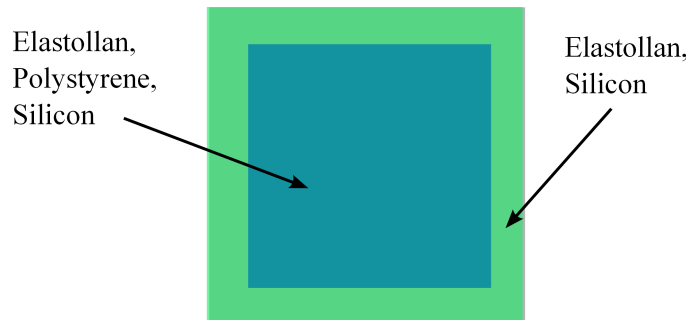


Fig. S2 Top view schematic of the final sample. The labels indicate the different material layers of the sample, listed in order from the top surface to the bottom surface. The ellipsometry measurements are performed in the region of Elastollan on silicon.

S2 Mechanical relationships for straining

Using Hooke's law¹, we can predict simple stress-strain relationships for our films under the assumption of Hookean elasticity. We assume that there is no stress acting in the direction orthogonal to the film (z -direction), i.e. $\sigma_z = 0$. We also know that the mechanical tension is related to stress through film thickness $T = h\sigma$. As such, we may derive a simple expression for isotropic tension generated upon straining:

$$T = \frac{Eh\varepsilon}{1-\nu}, \quad (\text{S1})$$

where ν is the Poisson ratio of the elastomer, which can be assumed to be 0.5, and E is the Young's modulus. Thus, εh is an appropriate indicator for the tension in our films. For the case of biaxial tension where the film is held fixed along one direction ($\varepsilon_{\text{low}} = 0$), we arrive at:

$$T_{\text{low}} = \frac{Eh\nu\varepsilon_{\text{high}}}{1-\nu^2}, \quad (\text{S2})$$

$$T_{\text{high}} = \frac{Eh\varepsilon_{\text{high}}}{1-\nu^2}, \quad (\text{S3})$$

where "high" and "low" indicate the high and low tension directions. Here, we see that under the assumption of Hookean elasticity, $T_{\text{high}} = 2T_{\text{low}}$, since $\nu = 0.5$. In the biaxial tension experiments, $\varepsilon_{\text{high}} \sim 100\%$, $h \sim 100$ nm, and $E \sim 10^7$ Pa², which leads to $T_{\text{high}} \sim 1.3$ N/m. Thus, it is clear that mechanical tension will dominate over interfacial tensions in determining the Laplace pressure in the rim.

Since volume is conserved in a material with $\nu = 0.5$, it is possible to predict the final thickness upon an isotropic strain:

$$h = \frac{h_0}{(1+\varepsilon)^2}, \quad (\text{S4})$$

as well as for a biaxial strain with $\varepsilon_{\text{low}} = 0$:

$$h = \frac{h_0}{(1+\varepsilon_{\text{high}})}. \quad (\text{S5})$$

Using the equations above, the initial film thickness h_0 was chosen to produce the desired h (50 nm or 100 nm) once strained.

S3 Equilibrium contact angle

To calculate the equilibrium contact angle that the rim would subtend with the substrate, we appeal to a balance of the interfacial and mechanical tensions depicted in Fig. S3. In this picture, we employ the simplifying assumption that the formation of holes does not alter the mechanical tension in the film. Carrying out the force balance in the horizontal direction yields:

$$T + \gamma_{\text{el},\nu} + \gamma_{\text{el},s} = (T + \gamma_{\text{el},\nu} + \gamma_{\text{el},l})\cos\theta_E + \gamma_{s,l}. \quad (\text{S6})$$

From the equation above, using the small angle approximation in which $\cos\theta \approx 1 - \theta^2/2$, it is straightforward to show that:

$$\theta_E = \frac{\theta_0}{\sqrt{1 + \frac{T}{\gamma_{\text{el},\nu} + \gamma_{\text{el},l}}}}. \quad (\text{S7})$$

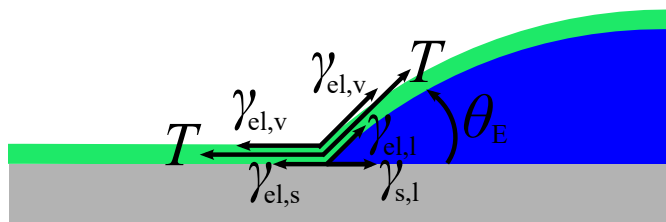


Fig. S3 Balance of interfacial and mechanical tensions to calculate the equilibrium contact angle that the rim subtends with the substrate.

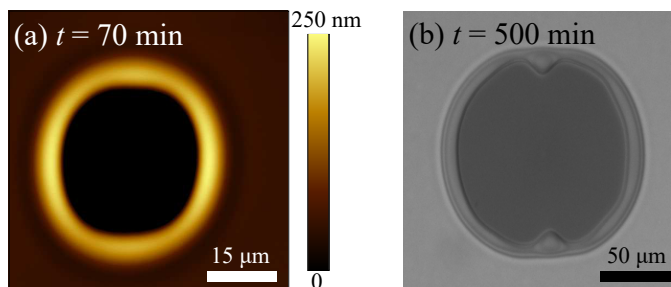


Fig. S4 (a) AFM scan of the hole from Fig. 3(b) once evidence of the rim instability sets in on the high tension sides of the hole. (b) Optical micrograph of the hole after the rim instability has started forming fingers at the high tension ends.

S4 Elongated holes at late times

As the elongated holes continue to grow beyond the early stages described in the main manuscript, we observe that the rim instability rapidly begins to set in on the high tension side of the rim. The early stages of this instability can be seen in an AFM scan of the hole in Fig. 3(b) at $t = 70$ min shown in Fig. S4(a) where there is a bulge forming in the high tension side of the rim. The later stage of this instability is showcased by the optical image in Fig. S4(b), where there are fingers forming at the high tension ends of the hole, yet the low tension side of the rim appears completely stable. In fact, the rim instability sets in at the high tension ends long before the stage of constant dewetting velocity has been reached. For this reason, the physics contained in Fig. 2(a), wherein holes exhibit a smaller v when the tension is larger, cannot be applied to these anisotropic experiments. We suspect that the root of this effect is that depending on a liquid cylinder's orientation, the Plateau-Rayleigh instability can become enhanced or suppressed when a tension anisotropy is introduced in the elastomeric film capping the liquid – this is beyond the scope of this study and will be investigated in future work.

S5 Supplemental movies

Movie S1 - Hole growth when the capping elastomer has an isotropic tension ($\epsilon h = 8$ nm). The movie is 750 min long.

Movie S2 - Growth of square holes.

References

- 1 S. Timoshenko and J. Goodier, *Theory of Elasticity, 2nd edition*, McGraw-Hill Book Company, Inc., New York, 1951.
- 2 H.-Y. Mi, X. Jing, M. R. Salick, W. C. Crone, X.-F. Peng and L.-S. Turng, *Adv. Polym. Tech.*, 2014, **33**, year.