Supporting Information for:

**Capillary Origami with Atomically Thin Membranes**

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1. Sample fabrication details

The monolayer MoS$_2$ films are directly grown on 1” circular fused silica substrates using the Metal-Organic Chemical Vapor Deposition.\textsuperscript{1} Patterning of the MoS$_2$ and Cu panels was performed directly on these substrates and proceeded according to the following recipe.

Patterning panels:
- Spin MMA (8.5) MAA 11 at 4000 rpm for 60 seconds
- Bake 170 C 1.5 min
- Spin PMMA 950K 4% at 4000 rpm for 60 seconds
- Bake 170 C 1.5 min
- Expose on contact aligner with 220 nm light, about 25 Joules per square centimeter (a one hour exposure on our contact aligner).
- Develop in IPA:MIBK 3 to 1 for 60 s
- Electron beam evaporate a 5 nm Ti adhesion layer and a 100 nm Cu layer.
- Lift-off by leaving the chip overnight in Microposit remover 1165. Then rinse in IPA.

Etching MoS$_2$
- Spin PMMA 495 4% at 4000 rpm for 60 seconds
- 60 s bake at 170 C
- Expose on contact aligner with 220 nm light, about 25 Joules per square centimeter (a one hour exposure on our contact aligner).
- Develop in IPA:MIBK 3 to 1 for 60 s
- Plasma etching
  - Oxygen plasma etch for 15 seconds at 50 W, 60 mTorr
  - SF$_6$ plasma etch for 10 seconds at 25 W, 30 mTorr
  - Oxygen plasma etch again for 15 seconds at 50 W, 60 mTorr
- Remove resist by leaving the chip overnight in Microposit Remover 1165. Then rinse in IPA.

2. Monolayer MoS$_2$ Characterization
We characterize our samples with AFM and Raman spectroscopy to confirm that the growth is complete and the samples are monolayer. Figure S1 shows a typical AFM image showing complete growth of the sample and a Raman spectrum with a peak separation of 19 cm\(^{-1}\), corresponding well to the expected value of 18 cm\(^{-1}\) for a monolayer sample.\(^2\)

Figure S1 – AFM and Raman spectrum of characteristic MoS\(_2\) samples. a. A typical AFM scan of a MoS\(_2\) sample before patterning. The image shows complete growth with slight overgrowth in a few regions. b. A Raman spectrum of a MoS\(_2\) sample after patterning. The peak separation between the E\(^{1\,2g}\) and the A\(_{1g}\) peaks is 19 cm\(^{-1}\), typical of monolayer MoS\(_2\).

3. Tensiometry measurement of Fluorinert FC-70 in DI water

Our experiments in this work used two liquids to form droplets in water: Fluorinert FC-70 and chloroform. While the surface tension of the chloroform-water interface can be found easily in the literature, few results exist for Fluorinert-water interfaces. We performed a simple pendant-drop tensiometry measurement on a droplet of Fluorinert in water without any MoS\(_2\) present. We imaged the drop with a home-built system consisting of a cuvette, a fiber-coupled light source, a zoom lens system, and a camera. By filling the cuvette with water and lowering a capillary glass tube filled with Fluorinert into the cuvette, we were able to image the resultant pendant drop.
The image was analyzed using a pendant drop software for ImageJ. This measurement gives a value of 42 ± 5 mN/m.

Figure S2 – Fluorinert FC-70 pendent drop. A micrograph of the image used to measure the surface tension of Fluorinert FC-70 in water.
4. Schematics of MoS$_2$ droplet wrapping

**Figure S3 – Schematics of folding of bare MoS$_2$.** Schematics of the folding patterns as a reference for the MoS$_2$ sheets shown in Figure 2a-c of the main text, showing that a. the square sheet folds into a packet resembling an envelope, b. the triangular sheet folds into a shape resembling a triangular pyramid, and c. the circular sheet folds into a semicircular “empanada” or “dumpling” shape.
5. Folding with graphene sheet

While we chose MoS$_2$ for this paper because it can be grown directly on fused silica substrates, this work could in principle be done with any two-dimensional material. To demonstrate this, Figure S2 shows a patterned circle of CVD graphene on a droplet of Fluorinert. We transferred CVD graphene to a fused silica cover slide and patterned it photolithographically, then released by undercutting the fused silica as with the MoS$_2$.

![Image](image1.png)

**Figure S4 -- Circular graphene sheet on Fluorinert droplet.** Image series showing a circle of CVD graphene wrapping a droplet of Fluorinert.

6. Calculation of wrinkle structure

In considering the expected wrinkle spacing and amplitude, we assume that the MoS$_2$ sheet is initially flat and corrugation-free on the droplet. Based on previous experimental works, as the droplet shrinks below a critical size for wrinkle formation, the number of wrinkles in a circular sheet should go as:

\[ n \sim 2\pi \left( \frac{\gamma W^2}{B} \right)^{1/4} \]

where $\gamma$ is the surface tension of the droplet, $W$ is the radius of the sheet, and $B$ is the sheet’s bending stiffness. Plugging in numbers for our system, $\gamma \approx 10 \text{ mN/m}$, $W = 10 \mu m$, $B \approx 10^{-16}$ J, we find that $n \sim 100$. This means that the wrinkles will be spaced by about 1 $\mu m$ around the
edge of the MoS$_2$ sheet. Since crumpling and folding dominate over wrinkles when the droplet radius $R$ is approximately equal to the circle radius $W$, we can estimate $R = W$ as the lower limit of droplet size for wrinkling behavior. This results in a compressive strain of approximately 10% at the edge of the sheet. As a result, each wrinkle must account for a length of $L/(10n)$. In our case, this yields a wrinkle amplitude of about 10 nm.

7. Chloroform microemulsion details

Microemulsions of chloroform were made from initially separated layers of one part chloroform to twenty parts deionized (DI) water. The emulsion was made by rapid pipetting to mix the solutions together and was added to the shape nets immediately after mixing to keep the chloroform and water from separating.

8. Folding of cubes with few-micron panels

While the few micron tip width of the capillary glass sets a minimum size for complete folding polyhedrons, self-folding with chloroform droplets permits folding of shapes a few microns in size.

Figure S5 – Folding of cube with 3 µm panels. Image time series showing the folding of a cube with 3 µm panels by a chloroform droplet.
References


