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Capillary Origami with Atomically Thin Membranes

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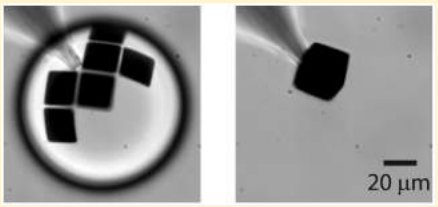
Supporting Information

ABSTRACT: Small-scale optical and mechanical components and machines require control over three-dimensional structure at the microscale. Inspired by the analogy between paper and two-dimensional materials, origami-style folding of atomically thin materials offers a promising approach for making microscale structures from the thinnest possible sheets. In this Letter, we show that a monolayer of molybdenum disulfide (MoS_2) can be folded into three-dimensional shapes by a technique called capillary origami, in which the surface tension of a droplet drives the folding of a thin sheet. We define shape nets by patterning rigid metal panels connected by MoS_2 hinges, allowing us to fold micron-scale polyhedrons. Finally, we demonstrate that these shapes can be folded in parallel without the use of micropipettes or microfluidics by means of a microemulsion of droplets that dissolves into the bulk solution to drive folding. These results demonstrate controllable folding of the thinnest possible materials using capillary origami and indicate a route forward for design and parallel fabrication of more complex three-dimensional micron-scale structures and machines.



KEYWORDS: *2D materials, origami, capillary, microstructures, MoS_2*

Although there is a robust toolkit for fabricating planar structures at the micron scale, making three-dimensional micron-scale objects is more challenging and requires a different set of tools. A growing body of work has demonstrated a variety of approaches for achieving three-dimensional geometries at the micron-scale, many of which rely on bending or folding originally planar structures.[1,2] There is significant interest in folding three-dimensional structures out of two-dimensional materials since they exist at the physical limit of thinness and can have a broad range of electronic and optical properties. Two-dimensional materials have recently been patterned, bent, and folded in a variety of ways to make three-dimensional shapes. Single-layer graphene has been patterned into stretchable, bendable graphene kirigami.[3] A variety of two-dimensional materials including MoS_2 have been rolled into nanoscrolls.[4−7] Layered stacks with graphene and silicon dioxide have been actuated with pH,[8] and layered stacks of graphene and polymer with temperature.[9]

One approach for folding that scales well for microscale structures is capillary origami, which drives folding with liquid surface tension. The way in which a droplet deforms and ultimately folds a flexible substrate has been studied from a variety of perspectives, both as a fundamental mechanics problem and as an engineering approach to fold specific shapes. Initial work on capillary origami demonstrated folding of a variety of shapes with polymer sheets and aqueous droplets on the millimeter scale.[10] Fundamental materials studies probed the way elastic sheets on droplets wrinkle,[11,12] as well as the wrapping dynamics of these systems.[13,14] Works akin to capillary origami achieved micro- and nanoscale bending and folding of shapes by patterning hinges of low-melting-temperature metals and heating to liquify them.[15−17] Others have performed capillary origami with water droplets at the 100-μm scale by using thin film ceramics to decrease bending stiffness.[18] Others still have shown control of capillary origami by electric and magnetic fields.[19,20] These structures have applications in optics[17,21] and drug delivery[22−24] from the nano- to macroscale. These works show that capillary origami is a powerful technique for folding small structures. In this work, we apply the technique of capillary origami to fold the

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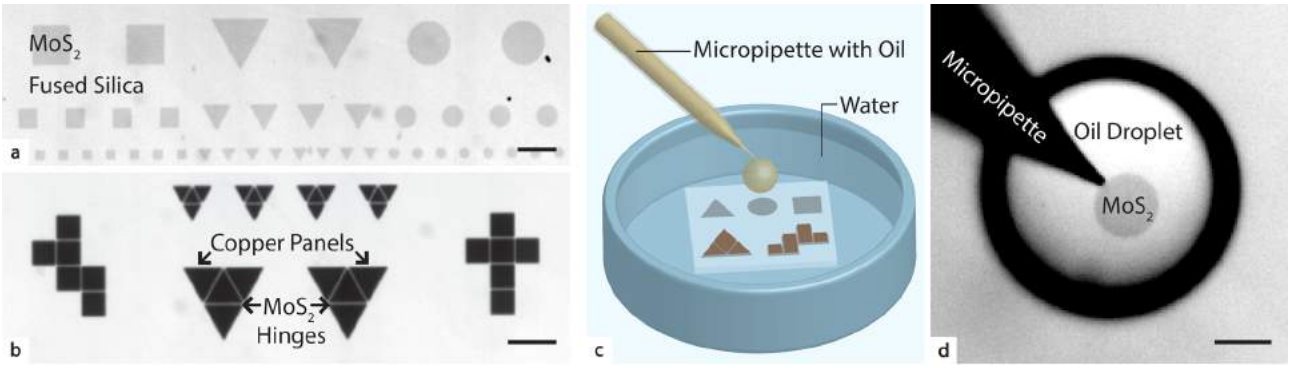

Figure 1. Sheets, shape nets, and experimental setup for folding. a, Patterned monolayer MoS_2 shapes and b, shape nets with Cu panels connected by MoS_2 hinges. The experimental setup, shown schematically in c, consists of these devices, released from the substrate with a brief hydrofluoric acid etch, in water with a micropipette filled with an oil attached to a pump for making droplets. A micrograph of the same setup is shown in d, with a MoS_2 circle picked up onto the surface of the droplet. All scale bars are 50 μm.

thinnest possible sheet, a two-dimensional material, by demonstrating surface-tension-induced folding of monolayer molybdenum disulfide (MoS_2) with droplets. This approach allows us to fold capillary origami with the thinnest sheets to date.

In capillary origami, the folding behavior is determined by comparing bending energy to surface energy. The characteristic length scale for capillary folding, known as the elastocapillary length, is defined as $L_{EC} = \sqrt{\frac{\kappa}{\gamma}}$, where κ is the two-dimensional bending stiffness of the sheet and γ is the surface tension of the droplet. This length scale approximately gives the radius of curvature to which a droplet will bend a rectangular sheet. Since the bending stiffness scales as the cube of the sheet thickness, $\kappa = \frac{Et^3}{12(1-\upsilon^2)}$, where E is the Young's modulus, t is the thickness, and υ is the Poisson ratio, atomically thin sheets such as MoS_2 are among the most flexible materials, permitting folding of micron-scale shapes in aqueous environments. For immiscible organics and oils in water, surface tensions are typically in the 10−100 mN/m range.[25] The Young's modulus of mono- and few-layer MoS_2 has been measured by nanoindentation and buckling methods to be approximately 250 GPa,[26−28] giving an expected bending stiffness for monolayer MoS_2 of approximately 10^{-17} J. The experimentally measured bending stiffness for freestanding graphene in aqueous solution is significantly stiffer than the expected value due to out-of-plane wrinkles, measuring in the range of 10^{-16}−10^{-15} J.[3] Because similar wrinkle structures could contribute in the case of MoS_2, the bending stiffness of our films is likely in the range of 10^{-17}−10^{-15} J. Using these ranges for surface tension and bending stiffness, we estimate that $L_{EC} \approx 10$−100 nm. This indicates that shapes down to 100 nm in size could in principle be folded using MoS_2 and oils or organic solvents. For MoS_2 sheets greater than about a micron in size, the surface tension of the droplet will easily dominate over the bending stiffness of the MoS_2, allowing us to fold micron-scale shapes with droplets in aqueous solutions.

We start with monolayer MoS_2 grown via metal−organic chemical−vapor deposition (MOCVD) on fused silica substrates (characterized by atomic force microscopy and Raman spectroscopy, Figure S1).[29] We photolithographically pattern the MoS_2 using a long, deep ultraviolet exposure of poly(methyl methacrylate) (PMMA), a resist chosen because it does not use water-based developers, which can cause delamination of the MoS_2 from the substrate. For devices with "shape nets", which consist of panels with MoS_2 hinges, we pattern copper panels with a thin (∼2 nm) titanium (Ti) adhesion layer on the MoS_2 via electron-beam deposition and liftoff. We then etch the MoS_2 into the desired shapes with a second layer of photolithography and an oxygen/sulfur hexafluoride (O_2/SF_6) plasma etch.

Figure 1a,b shows micrographs of bare MoS_2 shapes and of shape nets—copper (Cu) panels connected by MoS_2 hinges—respectively. Once the shapes are patterned, we release them via a substrate etch in 1:10 hydrofluoric acid in deionized water, with an etch time of seconds to a few minutes depending on the size of the shapes to be released. We rinse the chip and place it into pure deionized water, the environment in which further experiments take place. We then use a micropipette to place droplets of either Fluorinert FC-70 (3M), which has a surface tension in water of about 45 mN/m (Figure S2), or chloroform, which has a surface tension in water of 31.6 mN/m,[25] on the devices for micropipette folding and self-folding, respectively. For the micropipette folding, we apply pressure pulses to create the droplets with a Pneumatic PicoPump (WPI) connected to a micropipette with a 2-μm-diameter tip. Using micromanipulators, we bring a droplet into contact with the released MoS_2 and pick it up from the surface. Figure 1c schematically shows the chip, Petri dish, and micropipette, and Figure 1d shows a micrograph of a circle of patterned MoS_2 that has been picked up by a droplet on the micropipette.

Figure 2 (and Supporting Video 1) show the folding of both bare MoS_2 (a−c) and Cu-paneled shape nets (d,e). The folding takes place as Fluorinert flows back into the micropipette tip. Initially, for pressure P across a droplet of radius R, if $P \gtrsim \frac{-\gamma}{R}$, fluid flows into the micropipette in order to minimize the surface energy. When the droplet is completely wrapped by the MoS_2 or Cu/MoS_2, any further shrinking of the droplet does not decrease the surface energy of the system, but only further bends or crumples the MoS_2, which costs energy. The folding therefore stops when the wrapping is complete. Folding by this method takes anywhere from seconds to minutes and can be tuned by the initial droplet size and micropipette tip size, as well as by applying positive or negative pressure.

We observe that MoS_2 sheets fold to certain three-dimensional wrapped shapes based on their initial two-dimensional pattern. Squares and triangles generally fold

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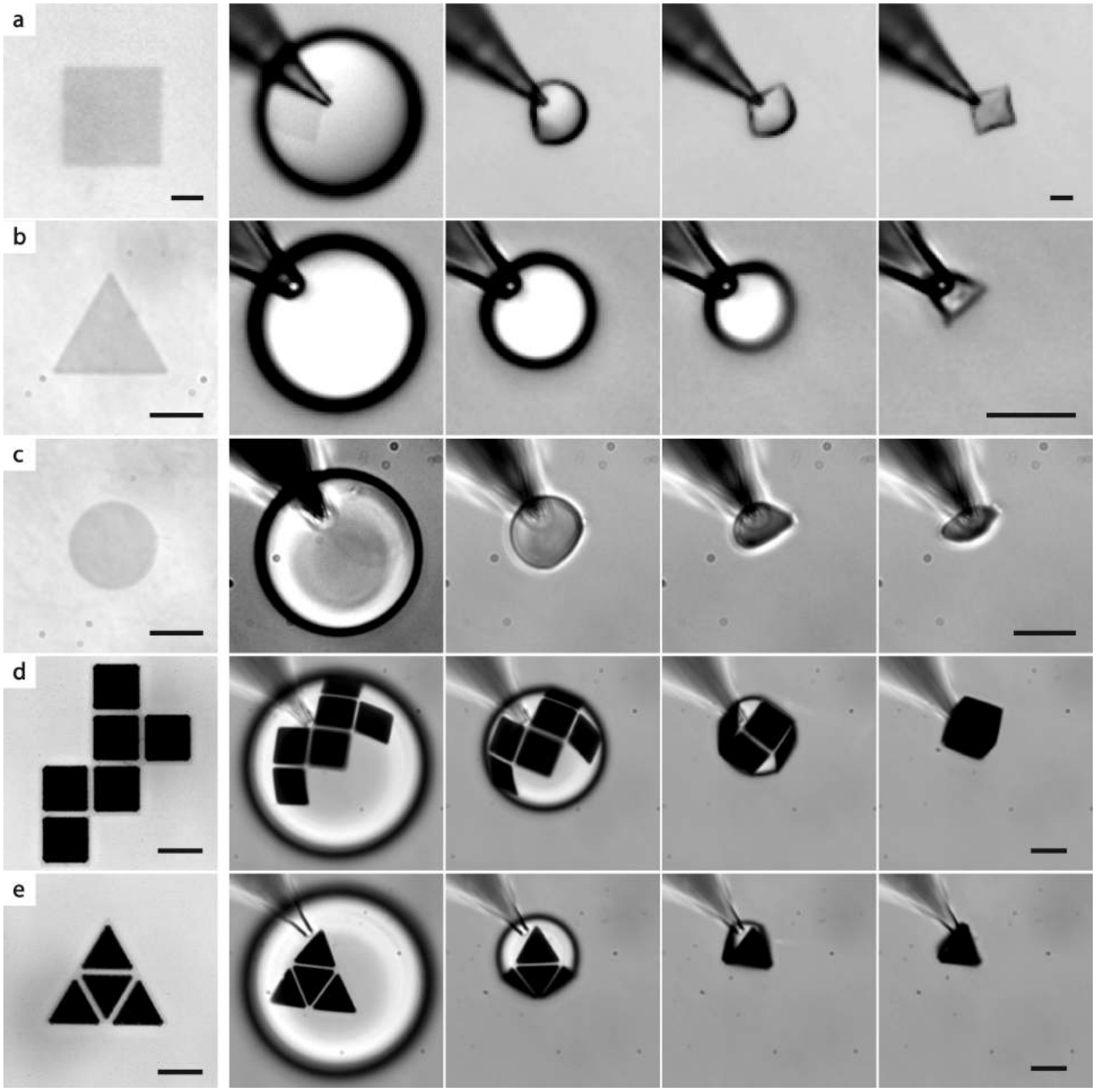

Figure 2. Image time series of the folding of both MoS_2 shapes and shape nets. The leftmost figure in each row shows the initial, unfolded shape. a–c, Square, triangular, and circular sheets folding into square, tetrahedron, and "empanada" packets, respectively. d,e, folding of shape nets (Cu panels and MoS_2 hinges) into a cube and a tetrahedron. All scale bars are 20 μm long. Videos of these folding shapes can be found in Supporting Video 1.

their vertices to center, such that square sheets make square packets and triangular sheets make tetrahedral packets, as shown in Figure 2a,b, respectively. Circular sheets of MoS_2 fold into both triangular and "empanada" packets (the latter of which is shown in Figure 2c), a phenomenon seen at a larger scale for circular polymer sheets in previous works.[13] To clarify the final shape of the packets, schematics of the folding behavior are shown in Figure S3. We also show that monolayer graphene sheets wrap Fluorinert in the same way (Figure S4). In experiments similar to ours, larger circular polymer sheets on droplets show wrinkle structures en route to folding.[11,12] Based on theory and these previous experiments,[12] we expect such wrinkles in our system to be around a micron in wavelength and 10 nm in amplitude (see SI 6 for calculation), making them difficult to detect by optical microscopy.

The addition of rigid panels to define MoS_2 hinges permits the folding of polyhedrons by design. Figure 2d,e show the folding of a cube and a tetrahedron from predesigned shape nets, respectively, each with panels that are approximately 20 μm across and hinges that are 2 μm wide. We designed the Cu panels to have a bending stiffness such that L_{EC} is greater than the size of the folded shape. This causes bending to take place preferentially at the MoS_2 hinge while leaving the panels undeformed. We select a panel thickness of 100 nm so that the panels have $\kappa \approx 10^{-11}$J and $L_{EC} \approx 10-100$ μm, suitable for folding polyhedra tens of microns in size. We pattern 2 μm MoS_2 hinges so that the bending stiffness of the hinge remains

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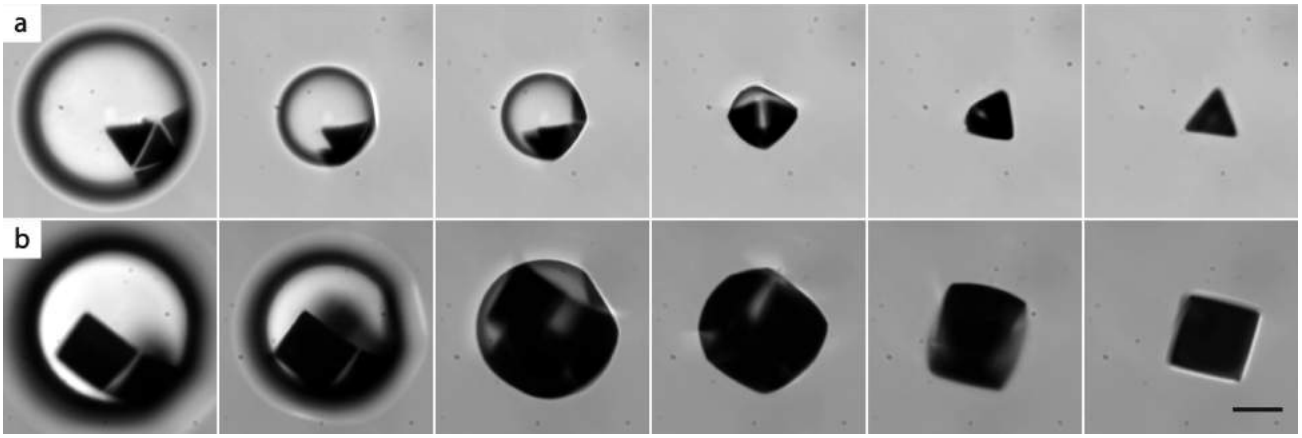

Figure 3. Self-folding with chloroform droplets. Image time series of self-folding of a, a tetrahedron, and b, a cube, with droplets of chloroform. Since chloroform is partially miscible in water, the shapes fold as the chloroform dissolves into the water. The scale bar is 25 μm long. Videos of these shapes can be found in Supporting Video 4.

negligible compared to the surface energy of the droplet. The shapes fold consistently provided that the hinges remained intact, and they can be re-expanded and refolded on the pipet tip without any apparent damage to the MoS_2 by gradually increasing and decreasing pressure (Supporting Video 2). They even survive being fired from the pipet tip with a pressure spike (Supporting Video 3).

Self-folding of polyhedrons can also be achieved by using organic solvents that are partially miscible in water. Figure 3a,b (and Supporting Video 4) show this process for a tetrahedron and a cube using chloroform, which has a solubility of 0.5% in water. We shoot droplets of chloroform onto the shapes individually by applying pressure pulses to a micropipette with a larger, 5 μm tip diameter. The shapes shown in Figure 3 start with droplets of approximately 50 μm in diameter and fold in 3 to 5 min, a time scale set by the dissolution rate of chloroform into water. While Figure 3 shows folding of shapes roughly 20 μm in size, we demonstrate folding of shapes down to 3 μm in size using this approach (Figure S5).

We use self-folding with chloroform droplets to fold many shapes simultaneously without a micropipette, an important step in moving this technique from a one-off demonstration to a fabrication method. An example is shown in Figure 4 (and Supporting Video 5). To achieve the folding of shapes in parallel, we add an emulsion of chloroform droplets to unreleased Cu/MoS_2 shape nets in deionized water. The shapes are left unreleased so that the process of adding the droplets does not scatter the shapes or cause one droplet to pick up many shape nets. Once the droplets settle on the shape nets, we add hydrofluoric acid to the solution to release the panels. As before, the droplets fold the shapes as chloroform dissolves into the bulk solution. The folding shown in Figure 4 takes place over about 15 min. This microemulsion approach to capillary origami makes possible massively parallel self-assembly of these microstructures.

In summary, we use capillary origami to fold atomically thin two-dimensional materials into micron-scale structures. These are the thinnest materials used for capillary folding to date. Further, we can fold these shapes in parallel, providing a route to the self-assembly of microstructures. In the future, development of techniques for folding these microstructures in air or for drying the structures subsequent to folding could increase the possible range of applications. The addition of

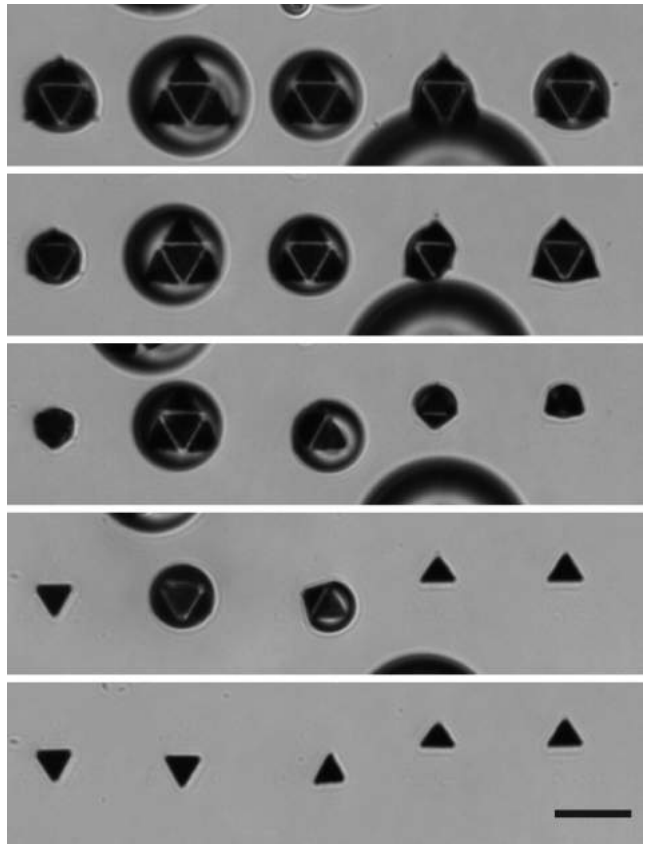

Figure 4. Parallel folding of shapes using chloroform emulsions. A microemulsion of chloroform was added to unreleased shape nets. Diluted hydrofluoric acid was then added to the solution so that the final ratio of hydrofluoric acid to water was approximately 1:50. Folding took place over about 15 min. The scale bar is 50 μm long. Video of this parallel self-folding can be found in Supporting Video 5.

active panels that include electronic, optical, or magnetic materials could permit the design of three-dimensional structures with functional properties. Applications in optics are particularly appealing based on the ability to controllably fold structures at the single micron-scale without the use of liquid metals, which can interfere with the designed optical components.[17] Furthermore, two-dimensional materials attaching various panels are a promising basis for a new class of smart

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surfactants that live at liquid interfaces, locally sensing their environment and responding to external stimuli.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.9b02281.

Sample fabrication details, monolayer MoS_2 characterization, tensiometry measurement of Fluorinert FC-70 in DI water, schematics of MoS_2 droplet wrapping, folding with a graphene sheets, calculation of wrinkle structure, chloroform microemulsion details, and folding of cubes with few-micron panels (PDF)

Folding MoS_2 sheets and shape nets (MP4)

Repeated folding (MP4)

Shooting a tetrahedron from the pipette tip (MP4)

Self folding with chloroform droplets (MP4)

Parallel self folding (MP4)

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Author Contributions

◆M.F.R. and K.L.M. contributed equally to this work. M.F.R., K.L.M., M.Z.M., I.C., and P.L.M. conceived the experiments. F.M., H.G., and K.K. performed the growth of the MoS_2 films under J.P.'s supervision. M.F.R. fabricated the samples. M.F.R., K.L.M., and M.A.W. performed the experiments with assistance from M.Z.M. and under P.L.M.'s and I.C.'s supervision. M.F.R., K.L.M., M.A.W., M.Z.M., and P.L.M. made the figures and wrote the manuscript with comments from all authors.

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Notes

The authors declare no competing financial interest.

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