Supporting Information

Adhesion of 2D MoS$_2$ to Graphite and Metal Substrates Measured by a Blister Test

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1. Growth and Characterization

We used chemical vapor deposition (CVD) to grow monolayer MoS\(_2\) flakes\(^{1,2}\) (Fig. S1a). Initially, MoS\(_2\) powder (Thermo Fisher Scientific, Molybdenum (IV) sulfide, 98%) is placed into the middle of the furnace inside an aluminum oxide crucible. A SiO\(_x\) wafer, which is cleaned with acetone, isopropyl alcohol (IPA), and deionized (DI) water and then exposed to ultraviolet (UV) for 5 minutes, is placed into a cooler region downstream of the MoS\(_2\) powder. Before starting the growth, the furnace is put under vacuum and purged with Argon (200 sccm) to remove air (specifically O\(_2\)). We then introduce 60 sccm Ar, 0.06 sccm O\(_2\), and 1.8 sccm H\(_2\) inside the tube. The growth process consists of three steps: (i) heating up to 900 °C for 15 minutes, (ii) holding at 900 °C for 15 minutes, and (iii) cooling the furnace to room temperature. During the growth, MoS\(_2\) powder sublimes and is carried downstream by Ar gas where it condenses onto the cooler SiO\(_x\) substrate.

After the growth, we observe numerous monolayer flakes in the optical microscope with ~100 \(\mu\)m side length. We deduce the approximate number of layers by using optical contrast\(^3\) and confirm this with both Raman spectroscopy (Fig. S1b) and photoluminescence (PL) spectroscopy (Fig. S1c). Both Raman and PL Spectroscopy were conducted in a Renishaw Raman InVia microscope using a 532 nm laser beam with 1200 l/mm gratings. In Raman spectroscopy, we obtained two prominent resonances\(^4\), the in-plane (E\(_{12g}\)) and out-of-plane (A\(_{1g}\)) vibrations located at 385 cm\(^{-1}\) and 405 cm\(^{-1}\) respectively, consistent with monolayer flakes. PL measurements\(^5\) show a peak at 1.9 eV (A exciton) and the absence of an indirect peak, both indicative of monolayer MoS\(_2\).
**Figure S1.** a) Schematics of the positions of the crucible and substrate in the furnace. b) Monolayer MoS$_2$ flakes on the SiO$_x$ substrate after the growth. c) Raman spectrum (Inset: Schematic of vibrational modes, E$_{12g}$ and A$_{1g}$). d) PL spectrum.
2. Metal Substrate Fabrication

Here we describe briefly the steps of the fabrication of the metal substrates. First, we clean the SiOₓ chips with acetone, IPA, and DI water. The surface of the SiOₓ is spin-coated with a photoresist solution of S1818 at 2500 rpm, and the substrate is put on a hot plate at 115 °C for 1 minute. We use a contact aligner with a mask with 5 μm diameter circles on it to create the photoresist pattern. The spin-coated chips are exposed to UV for 20 seconds with 8 mW power. The parts which are exposed to UV are removed with MF-319 Developer and then SiOₓ and Si are etched by Reactive-Ion-Etching (RIE). We use 3.1 sccm O₂ and 25 sccm CF₄ at 100 mTorr pressure and 150 W power for 13 minutes of etching. This creates wells with a depth between ~ 600-900 nm. In the final step, we remove the photoresist, in a bath of 1165 Remover where the chip soaks for 12 hours at 110 °C. To further clean the chips of photoresist residue, we put the etched wells into an O₂ plasma at 300 Watts with 300 sccm of O₂ for 2 minutes. The microcavities are then placed into a thermal evaporator. Germanium, Chromium, and Titanium are evaporated over the etched well with a thickness of 300 Å and a rate of 0.5 Å/s. For the gold substrate, 180 Å-thick Titanium is used as an adhesive and 450 Å of gold is subsequently evaporated. A similar process is utilized for the other metals studied (Fig. S2).
Figure S2. Microfabrication of the metal wells.
3. Graphite Substrate Fabrication

The preparation of the graphite wells starts with cleaning the SiO_x surface with acetone, IPA, and DI water. With the 'Scotch-Tape' method, freshly exfoliated flakes on tape are pressed against the oxide surface. The tape is peeled from the surface very slowly (~1 mm/min). The substrate is then spin-coated with S1818 at 2500 rpm and kept on the hot plate at 115 °C for 1 minute. The spin-coated chips are exposed to UV for 20 seconds with 8 mW power. Again, we use the same patterned mask during UV exposure to form the circular wells. Using RIE, we etch the wells to ~600-900 nm deep by deploying the same etching parameters as described above for the metal devices. Devices are then placed into the Remover 1165 bath at 110 °C over 12 hours and then exposed to O_2 plasma to remove any remaining photoresist (Fig. S3).
Figure S3. Microfabrication of the graphite wells.
4. Graphite Surface Treatment

Due to the O$_2$ plasma exposure for cleaning the graphite surface after photolithography, we performed Raman spectroscopy to determine if any surface chemistry changes took place. First, we carried out Raman spectroscopy over the freshly exfoliated flake (Fig. S4a) and again after photolithography and exposure to O$_2$ plasma during the final cleaning step (Fig. S4b). As can be seen in Figure S4b, a small D peak is introduced after O$_2$ plasma exposure indicative of the formation of defects$^6$ or graphite oxide formation$^7$ in the graphite lattice. However, the small size of the D peak suggests that the surface is not heavily oxidized.

Figure S4. a) Optical image and Raman Spectrum of the freshly exfoliated graphite flake. Scale bar is 30 µm. b) Optical image and Raman spectroscopy of the etched FLG wells which exposed to O$_2$ plasma. The peak at 1400 cm$^{-1}$ due to defects. Scale bars are 20 µm.
5. MoS$_2$ Transfer Procedure

The transfer of MoS$_2$ begins by spin coating the CVD-grown MoS$_2$ flakes with PMMA at 2500 rpm. We create a window on the thermal-release tape and it is stamped onto the PMMA-covered MoS$_2$ substrate. Utilizing the hydrophilic nature of the SiO$_x$/MoS$_2$ interface, we put this combination into water and let the water separate the MoS$_2$ flakes from the SiO$_x$ substrate. This leaves us with a MoS$_2$/PMMA/Thermal-Release-Tape (MPT) combination.

Following the preparation of the MPT combination, metal and graphite substrates are both placed on a hot plate at 85 °C, and MPT is put onto the target substrate. For the graphite well, the transfer is carried out under the optical microscope with a custom-made apparatus that is used to keep the MPT still while approaching to surface. The Thermal-Release-Tape is peeled off from the surface easily with heat and the MoS$_2$/PMMA then sticks to the substrate. Before the annealing process, we put this device into a desiccator overnight (< 12 hr) to avoid any damage that trapped air inside the cavity may cause. The annealing process is carried out at 350 °C for 7 hours and under 20 sccm of Argon flow to help remove the PMMA from the surface (Fig. S5a). The transfer results are shown in the Fig. S5b (graphite) and Fig. S5c (gold).
**Figure S5.** a) Schematic of monolayer MoS$_2$ flakes transfer over the target substrate. b-c) Optical image and schematic of the final result of the MoS$_2$ transfer over the graphite well and gold wells respectively. The dashed line in (c) shows the boundary of the MoS$_2$ flake.
6. Deriving the Expression for Pressure Difference and Deflection

Following the Hencky’s solution for the deformation of a pressurized clamped axisymmetric membrane\(^8,9\), we obtain the deflection profile as an infinite summation of even powers of normalized radius;

\[
z(r) = a \left( \frac{\Delta p a}{Et} \right)^{1/3} \sum_{n=0}^{\infty} A_{2n} \left( 1 - \left( \frac{r}{a} \right)^{2n+2} \right)
\]

with \(A_0 = 1/B_0\), \(A_2 = 1/2B_0^2\), \(A_4 = 5/9B_0^3\), \(A_6 = 55/72B_0^4\), \(A_8 = 7/6B_0^5\), and so on. \(E\) is the bulk Young’s modulus, \(t\) is the thickness of the membrane, and \(\Delta p\) is the pressure difference. The maximum height of the blister at the center can be found by \(\delta \equiv z(r = 0)\) which yields:

\[
\delta = a \left( \frac{\Delta p a}{Et} \right)^{1/3} \sum_{n=0}^{\infty} A_{2n}
\]

By setting \(K(v) = 1/\left( \sum_{n=0}^{\infty} A_{2n} \right)^3\) and rearranging Eqn. S6.2, the final expression results in Eqn.1 of the main text.

One assumption of the constant-N blister test is the expansion of the membrane occurs isothermally, so we can model the change in the volume by ideal gas law which gives \(p_0 V_0 = p_{int}(V_0 + V_b)\), where \(V_0\) is the volume of the microcavity and \(\Delta p = p_{int} - p_{ext}\). From Hencky’s solution, the volume under the bulge is determined by the formula \(V_b = C(v)\pi a^2 \delta\) where \(C(v = 0.29) = 0.552\) since \(v = 0.29\)\(^9,10\) for MoS\(_2\).
Below, we tabulate the constants $C, K$ for a range of MoS$_2$ Poisson’s ratio values commonly found in the literature (Table S1) and the corresponding calculated 2D Young’s modulus and work of separation using these values. As can be seen, it does not influence the work of separation and has only a minor effect on the calculated elastic modulus.

<table>
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<tr>
<th>Device ID:</th>
<th>Poisson's Ratio ($\nu$)</th>
<th>K($\nu$)</th>
<th>C($\nu$)</th>
<th>$E_{2D}$ (N/m)</th>
<th>$\Gamma_{sep}$ (J/m$^2$)</th>
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<td>R13</td>
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<td>0.32</td>
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<td>0.25$^{11}$</td>
<td>3.39</td>
<td>0.523</td>
<td>228.3</td>
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Table S1. Values for the constants C and K obtained with different Poisson’s ratios and the $E_{2D}$ and $\Gamma_{sep}$ calculated using the corresponding constants.
7. Determining the Young’s Modulus of MoS$_2$ over Graphite Devices

We examined 44 devices of MoS$_2$ over graphite substrates. In Fig. S6a, we plot $K(\nu) \delta^3/\alpha^4$ vs $\Delta p$ for 8 representative devices. In Fig. S6b, we show the full data set of the calculated E$_{2D}$ values which were used to determine the work of separation (Fig. 4a in main text). Figure S6c shows an optical image of the graphite devices pinned to their initial radii, while the nearby membranes over the SiO$_x$ wells show delamination at the same input pressure. Figure S6d-e are AFM images of those same devices showing that the MoS$_2$ over the graphite wells remains adhered while those on the SiO$_x$ are delaminated for the same input pressure confirming the lower work of separation for MoS$_2$/SiO$_x$. 
Figure S6. a) $K(n)\delta/a^4$ vs $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits used to determine $E_{2D}$ of each device. b) $E_{2D}$ for each device. c) Optical image of MoS$_2$ over the graphite and SiO$_x$ substrates (Device A is on the graphite and Devices B, C, and D are on the SiO$_x$). d) 3D AFM image of the devices labeled at (c). e) AFM Amplitude image of the devices labeled in (c).
8. Using Photoluminescence to Verify the Clamping Condition Assumed in Hencky’s Model

MoS$_2$ shows changes in the optical band gap of $\sim$ 100 meV/\% for biaxial strain$^{1,12}$. Our membranes are subjected to biaxial strain near the center via a pressure difference $\Delta p = p_{int} - p_{ext}$. To validate the assumption in the Hencky model of a perfectly clamped circular membrane we used strain measurements determined by PL measurements and correlated them with strain measurements made by the AFM. A line scan of a series of PL measurements over the blister is shown in Fig. S7a. These are used to accurately determine the strain at the center of the membrane. In Figure S7b, we plot the PL response that corresponds to the points labeled with different colors indicating the different points (Fig. S7c, and d). In addition, we fit the Voigt function to each spectrum to follow the A exciton peak position$^{13}$. We repeated the PL measurements for every new input pressure until delamination, and AFM scans accompany those measurements. We then correlated the AFM scans and PL measurements$^{8,9,14}$. We use the stress-strain governing equations

$$\sigma_r - \nu \sigma_\theta = E t \varepsilon_r \quad \text{S8.1}$$

$$\sigma_\theta - \nu \sigma_r = E t \varepsilon_\theta \quad \text{S8.2}$$

where $\sigma_r$ and $\sigma_\theta$ are radial and tangential membrane stresses respectively and $\varepsilon_r$ and $\varepsilon_\theta$ are the radial and tangential strains. At the center of the membrane where $r = 0$, $\varepsilon_\theta = \varepsilon_r$ which is equivalent to perfect biaxial strain ($\varepsilon_b$). We can also write the equation for $\sigma_r$ as an infinite series of even powers of radius $r$,

$$\sigma_r = \left(\frac{E t \Delta p^2 a^2}{64}\right)^{1/3} \sum_{n=0}^{\infty} B_{2n} \left(\frac{r}{a}\right)^{2n} \quad \text{S8.3}$$

with $B_2 = -1/B_0^2$, $B_4 = -2/3B_0^5$, $B_6 = -13/18B_0^8$, $B_8 = -17/18B_0^{11}$, etc.
If we combine Eqn. S6.2 and S8.3 and Eqn. 1 from the main text, the final expression yields:

$$\varepsilon_b = \frac{B_0(1 - \nu)K(\nu)^{2/3}}{4} \left(\frac{\delta}{a}\right)^2$$

S8.4

where $B_0$ and $K$ depend on Poisson’s ratio and $K(\nu) = \frac{1}{\left(\sum_0^\infty A_{2n}\right)^3}$. Using the Poisson’s ratio for MoS$_2$ ($\nu = 0.29$), we obtain $B_0 = 1.72$ and $K = 3.54$. We then substitute these values back into Eqn. S8.4 to obtain:

$$\varepsilon_b = 0.709 \left(\frac{\delta}{a}\right)^2$$

S8.5

From this expression, and knowing the maximum deflection and radius of the blister we calculate the biaxial strain at the center.

In Figure S7e, we plot the PL at the center of the blister vs. the strain calculated from the corresponding AFM scan for MoS$_2$ over the gold wells. The change in strain agrees with the theoretical change (~ 100 meV/%) validating the assumption of a perfectly clamped circular membrane assumed in the Hencky model. The laser is focused to a diffraction limited spot size of ~ 0.5 µm. The lateral spatial resolution is therefore ~ 0.5 µm. During the PL measurement, laser is kept focused over the scanned area.
Figure S7. a) AFM image of the blister (height channel). The dashed line passes through the center of the blister where we conduct PL measurement over the points which are located along both sides of the blister. The scale bar is 2 μm. b) Positions of the PL measurement point on the cross-section of the device in (a). c) PL response of the data points labeled in (b) which we use the peak points in strain calculation. Solid lines correspond to points from the left edge of the blister to the center of the blister and dashed-dotted lines from the center to the right edge blister. d) Corresponding biaxial strain of the points in (c) by using their PL measurements. e) Calculation of the strain using the AFM results of each pressure change and matching them with corresponding PL data of MoS$_2$ on the gold wells. The dashed line is the theoretical change of the A exciton with respect to strain$^1$. 
9. Strain Energy Derivation

In the paper, we closely follow the Wan et al. study\textsuperscript{14,15} to calculate the contribution of the strain energy to our free energy model. First, we utilize equations described in the Hencky’s solution.

Aside from the governing equations mentioned before (Eqn. S6.1, S6.2, S8.1, and S8.2), we also use the following equations:

\[ \sigma_\theta = \frac{d}{d r}(r \sigma_r) \]  \hfill \text{(S9.1)}

\[ \epsilon_\theta = \frac{u}{r} \]  \hfill \text{(S9.2)}

where \( u \) is radial displacement and \( r \) is radial polar coordinate. Wan et al. showed that the strain energy in the membrane is equal to the work done by the external load, i.e., gas pressure while keeping the blister radius \( a \) fixed.

\[ F_{\text{mem}} = \int \Delta p(\delta) \, dV(\delta) \bigg|_{a} = \frac{\Delta p \, V_b}{4} \]  \hfill \text{(S9.3)}

Assuming \( \xi = \frac{r}{a} \), we obtain the volume of the bulge as,

\[ V_b = \int_0^a z(r) \, 2 \pi r \, dr = a^2 \left( \frac{\Delta p \, a^x}{Et} \right)^{\frac{1}{3}} \int_0^1 z(\xi)2\pi \xi d\xi \]  \hfill \text{(S9.4)}

Using S6.1, we get:

\[ F_{\text{mem}} = \frac{\Delta p \, a^2}{16 \left( \frac{\Delta p \, a^4}{Et} \right)^{\frac{1}{3}}} \left( \frac{\pi}{8B_0} + \frac{\pi}{12B_0^4} + \frac{5\pi}{48B_0^7} + \frac{11\pi}{72B_0^{10}} + \frac{35\pi}{144B_0^{13}} + \frac{205\pi}{504B_0^{16}} + \frac{17051\pi}{24192B_0^{19}} + \frac{2864485\pi}{2286144B_0^{22}} + \frac{20772653\pi}{9144576B_0^{25}} + \frac{135239915\pi}{32332608B_0^{28}} + \frac{42367613873\pi}{5431878144B_0^{31}} + \ldots \right) \]  \hfill \text{(S9.5)}
It can be shown that Eqn. S9.5 is equal to the conventional expression for membrane strain energy in Eqn. S9.6.

\[ F_{\text{mem}} = \int_{0}^{a} \left( \frac{1}{2} \sigma r \epsilon r + \frac{1}{2} \sigma \theta \epsilon \theta \right) 2\pi r \, dr \]  
S9.6

We obtain \( \epsilon r \) and \( \epsilon \theta \) by manipulating S8.1 and S8.2, respectively. After defining non-dimensional tangential stress component \( \sigma \theta = \frac{d}{dr}(r \sigma r) \Rightarrow \sigma \theta = \frac{d}{d\xi}(\xi \sigma r) \), if we plug these back into Eqn. S9.6, we get:

\begin{align*}
F_{\text{mem}} &= \pi a^2 \int_{0}^{1} \left( \sigma^2 r + \sigma^2 \theta - 2v \sigma r \sigma \theta \right) \xi d\xi \\
&= \pi a^2 \int_{0}^{1} \left( 2(1 - v) \sigma^2 r + 2(1 - v) \xi \frac{d\sigma r}{d\xi} + \xi \left( \frac{d^2 \sigma r}{d\xi^2} \right) \right) \xi d\xi \\
&= \pi a^2 \int_{0}^{1} \left( d \left( 1 - v \right) \sigma^2 r \xi^2 \sigma r + \xi^3 \left( \frac{d^2 \sigma r}{d\xi^2} \right) \right) d\xi
\end{align*}
S9.7

The clamped boundary condition gives us the following relation:

\[ u(\xi = 1) = 0 \Rightarrow \sigma \theta(\xi = 1) - v \sigma r(\xi = 1) = 0 \Rightarrow \]

\[ (1 - v) \sigma r(\xi = 1)^2 = -\xi \sigma r(\xi = 1) \left( \frac{d\sigma r}{d\xi}(\xi = 1) \right) \]  
S9.8

After integrating the first term in the integrand, the final formula yields:

\[ F_{\text{mem}} = \pi a^2 \left( \int_{0}^{1} \xi^3 \left( \frac{d\sigma r}{d\xi} \right)^2 \right) d\xi - \pi \left( \sigma r(\xi = 1) \frac{d\sigma r(\xi = 1)}{d\xi} \right) \]  
S9.9
Computing the Eqn. S9.9 using the series expressions for $\sigma_r$ (Eqn. S8.3) gives us:

$$ F_{mem} = \frac{\Delta p}{16} a^2 \left( \frac{\Delta p}{Et} \right)^{\frac{1}{3}} \left[ \frac{\pi}{8B_0} + \frac{\pi}{12B_0^3} + \frac{5\pi}{48B_0^5} + \frac{11\pi}{72B_0^7} + \frac{35\pi}{144B_0^9} + \frac{205\pi}{504B_0^{11}} + \frac{17051\pi}{24192B_0^{13}} + \frac{2864485\pi}{2286144B_0^{15}} + \ldots \right] S9.1 $$

As can be seen, the series in Eqns. S9.5 and S9.10 are identical.

10. Relamination of MoS$_2$ over the Metal and Graphite Wells

In Figure S8, the complete deflation data for 9 devices of MoS$_2$ over gold substrates are shown. We obtain a mean value of $\Gamma_{adh} = 0.017 \pm 0.005$ J/m$^2$. Figure S8b is the optical image of the delaminated device, and Figure S8c-h are AFM images showing relamination. The adhesion hysteresis observed is reminiscent of AFM based nanoindentation studies. In these studies the hysteresis has been attributed to moisture$^{17,18}$, viscoelasticity$^{19}$, plasticity$^{20}$, and surface instabilities$^{21,22}$. Similar mechanisms as these may be at work in our case as well. The
measurements of the deflection and radius are performed along the cross-sections shown in Fig. S8c and only the mean values of these measurements are shown in Fig. S8a for clarity.

**Figure S8.** a) Complete data of deflation for 9 devices. Each color represents a different device. The dashed line is the mean value of all samples ($\Gamma_{adh} = 0.017 \pm 0.005 \text{ J/m}^2$ (Dashed line). b) Optical image of the delaminated devices. The scale bar is 10 $\mu$m. c-d) AFM images (amplitude channel) of the device in (b). Elapsed times are put on top of each photo. The dashed lines in (c) are the cross-sections that are used for deflection and radius change. The scale bars are 3 $\mu$m.

In Figure S9, we plot the deflation data sets for the MoS$_2$ on the FLG substrates. We performed the measurements on 9 different devices and found $\Gamma_{adh} = 0.057 \pm 0.008 \text{ J/m}^2$ (dashed line). Fig. S9b is the optical image of the devices and its deflation behavior is shown in Fig. S9c-k with each labeled with the elapsed time of the measurement.
Figure S9. a) Complete deflation data for 9 graphite devices. Each color represents a different device. The dashed line is the mean value of all samples shown in Fig. 4d ($\Gamma_{\text{rel}} = 0.057 \pm 0.008 \text{ J/m}^2$). b) Optical image of a delaminated device. The scale bar is 5 $\mu$m. c-k) AFM images (amplitude channel) of the device in (b). Elapsed times are included. The dashed lines in (c) are the cross-sections that are used for the measurement of deflection and radius. The scale bars are 3 $\mu$m.

In Figure S10, we examine the MoS$_2$ over Si wells and in Fig. S10a we plot $K(v)\delta^3/a^4$ vs $\Delta p$. A linear fit to the data is used to determine $E_{2D}$ for each device. These calculated $E_{2D}$ values are used in the separation and adhesion energy calculations of these membrane from the Si wells. In Figure S10b, we show the values of $E_{2D}$ determined from the fit in Fig. S10a. Figure S10c is the deflation data for the MoS$_2$ on the Si substrates. We performed the measurements on 5 different devices. We found a work of adhesion $\Gamma_{\text{adh}} = 0.03 \pm 0.0018 \text{ J/m}^2$ (dashed line).
Figure S10. Data for Si substrates. a) $K(v)\delta/\alpha^4$ vs $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits that are used to calculate $E_{2D}$ of each device. Symbols are color-coded. b) $E_{2D}$ for each device c) Maximum deflection, $\delta$, and radius during the deflation. The dashed line is the mean value of all samples.

In Figure S11 we examine the MoS$_2$ over SiO$_x$ wells and in Fig. S11a we plot $K(v)\delta/\alpha^4$ vs $\Delta p$. A linear fit to the data is used to determine $E_{2D}$ for each device. In Figure S11b, we show the values of $E_{2D}$ determined from the fit in Fig. S11a. These calculated $E_{2D}$ values are used in the separation and adhesion energy calculations of these membrane from the SiO$_x$ wells.
Figure S11. Data for SiO$_x$ substrates. a) $K(\nu)\delta^3/a^d$ vs $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits that are used to calculate $E_{2D}$ of each device. Symbols are color-coded. b) $E_{2D}$ for each device.

In Figures S12 through S14, we examine the MoS$_2$ over Titanium, Chromium, and Germanium wells. In each figure, we plot $K(\nu)\delta^3/a^d$ vs $\Delta p$. A linear fit to the data is used to determine $E_{2D}$ for each device. Later, we show the values of $E_{2D}$ determined from the fits which are used in the separation and adhesion energy calculations for each substrate. The average work of adhesion can be found in the caption of the figures for each material.
Figure S12. Data for Titanium substrates. a) $K(v)\delta/a^4$ vs $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits that are used to calculate $E_{2D}$ of each device. Symbols are color-coded. b) $E_{2D}$ for each device. c) $\delta$ vs radius during deflation. The dashed line is the mean value of all samples. $\Gamma_{\text{adh}} = 0.03 \pm 0.004$ J/m$^2$. 
Figure S13. Data for Chromium substrates. a) $K(v)\delta/a^4$ vs $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits that are used to calculate $E_{2D}$ of each device. Symbols are color-coded. b) $E_{2D}$ for each device. c) $\delta$ and radius during deflation. The dashed line is the mean value of all samples. $\Gamma_{adh} = 0.036 \pm 0.018$ J/m$^2$. 
Figure S14. Data for Germanium substrates. a) $K(v)\delta^4/\alpha^4$ against $\Delta p$ for CVD-grown MoS$_2$ membranes. Dashed lines are the linear fits that are used to calculate $E_{2D}$ of each device. Symbols are color-coded. b) $E_{2D}$ for each device. c) $\delta$ and radius during deflation. The dashed line is the mean value of all samples. $\Gamma_{adh} = 0.012 \pm 0.002$ J/m$^2$.

11. Roughness Measurements over the Substrates

To determine the influence of surface roughness on the work of separation, we measured the surface roughness of our substrates using an AFM. First, we performed the AFM scans after the fabrication of the substrate. Following the MoS$_2$ transfers over the wells, we performed another AFM scan over the MoS$_2$ covered areas. AFM scans are conducted in a 250 nm x 500 nm area in
tapping mode. Then, we analyzed 100 nm x 100 nm sub-areas within the scans to determine the root-mean-square (rms) values of the roughness using the NanoScope Analysis 2.0 program. The same scan sizes, and parameters are used for all of the scans.

In the following graphs, we show the AFM height images and the representative line cut through the scan area to demonstrate the surface roughness for each substrate used in our experiment.

Figure S15. Si substrate. a) AFM scan over bare substrate fabrication of wells. b) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
Figure S16. SiO$_x$ substrate. a) AFM scan over bare substrate after fabrication of wells. b) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
**Figure S17.** Graphite substrate. a) AFM scan after fabrication of wells. b) Scan over bare substrate after annealing. c) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
Figure S18. Gold substrate. a) AFM scan after fabrication of wells. b) Scan over bare substrate after annealing. c) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
**Figure S19.** Titanium substrate.  

a) AFM scan after fabrication of wells  
b) Scan over bare substrate after annealing.  
c) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
Figure S20. Chromium substrate. a) AFM scan after fabrication of wells b) Scan over bare substrate after annealing. c) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
Figure S21. Germanium substrate. a) AFM scan after fabrication of wells b) Scan over bare substrate after annealing. c) Scan over MoS$_2$ covered area after annealing. Scale bars are 50 nm.
Figure S22. Aluminium substrate. a) AFM scan after fabrication of wells. Scale bar is 50 nm.

In Table S2, the results of the AFM scans over the substrates are tabulated.

<table>
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<tr>
<th>Substrate Material</th>
<th>Roughness Measurements (Averaged RMS Values)</th>
<th>Work of Separation (J m⁻²)</th>
<th>Work of Adhesion (J m⁻²)</th>
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</thead>
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<tr>
<td></td>
<td>Over the Bare Substrate Before Annealing (nm)</td>
<td>Over the Bare Substrate After Annealing (nm)</td>
<td>Over the MoS₂ Flake (nm)</td>
</tr>
<tr>
<td>Si</td>
<td>0.15±0.02</td>
<td>N/A</td>
<td>0.21±0.04</td>
</tr>
<tr>
<td>SiO₂</td>
<td>0.18±0.01</td>
<td>N/A</td>
<td>0.20±0.02</td>
</tr>
<tr>
<td>Graphite</td>
<td>0.21±0.04</td>
<td>0.27±0.03</td>
<td>0.26±0.01</td>
</tr>
<tr>
<td>Gold</td>
<td>0.7±0.08</td>
<td>0.57±0.04</td>
<td>0.6±0.02</td>
</tr>
<tr>
<td>Chromium</td>
<td>0.48±0.02</td>
<td>0.45±0.03</td>
<td>0.5±0.04</td>
</tr>
<tr>
<td>Germanium</td>
<td>0.32±0.04</td>
<td>0.32±0.01</td>
<td>0.33±0.02</td>
</tr>
<tr>
<td>Titanium</td>
<td>0.4±0.08</td>
<td>0.35±0.02</td>
<td>0.36±0.1</td>
</tr>
<tr>
<td>Aluminium</td>
<td>3.8±0.8</td>
<td>N/A</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Table S2. Summary of roughness measurements.

12. Transfer of MoS₂ Membranes over Various Substrates
Videos showing the transfer of the MoS$_2$ onto the Silicon Oxide (video 1), Chromium (video 2), Titanium (video 3), and Aluminum (video 4) surfaces. In video 4, we attempted to transfer MoS$_2$ over the aluminum coated surface but were unsuccessful. Aluminum had the highest surface roughness and MoS$_2$ was unable to be transferred.

References


(22) Schwartz, L. W.; Garoff, S. Contact Angle Hysteresis on Heterogeneous Surfaces.