– Supporting Information –

Tuning Optical Signatures of Single- and Few-Layer MoS_2 by Blown-Bubble Bulge Straining up to Fracture

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S1. Hencky's Model

In this section, we show the estimation of pressure-induced deflection and strain profiles in the bulging PDMS using Hencky's model.[1] Hencky's model is a series of analytical solutions for calculating the deformation and strain of a pressurized circular diaphragm with fixed boundary at its edge. In our experiments, a PDMS film is inflated with circular clamping, hence it closely matches with Hencky's case. When pressure difference is applied across the thin film, the film bulges up or down depending on the polarity of pressure difference. The deformation of bulging can be decomposed to radial and out-of-plane displacements, which are noted as u and w, respectively, with respect to dimensionless loading of $q = \Delta pa / E_{\mathrm{Y}} t$, where Δp, a, E_{Y}, and t are pressure difference, radius, Young's modulus and thickness of the circular diaphragm, respectively. The resulting bulging induced strains in the film are

$$\varepsilon_\theta = \frac{U}{\rho}, \tag{S1}$$

$$\varepsilon_r = \frac{dU}{d\rho} + \frac{1}{2}\left(\frac{dW}{d\rho}\right)^2, \tag{S2}$$

where ε_θ is tangential strain, ε_r is radial stain, W is dimensionless out-of-plane displacement, U is dimensionless radial displacement, and ρ is dimensionless radial coordinate, respectively. The dimensionless parameters are attained by normalizing each parameter using the radius of the diaphragm, (*e.g.*, $W=w/a$). The governing equations at equilibrium are given by balancing pressure loading and stresses in the diaphragm:

$$\sigma_\theta = \frac{d}{d\rho}\left(\rho\sigma_r\right), \tag{S3}$$

$$\sigma_r \frac{dW}{d\rho} = -\rho\frac{q}{2}, \tag{S4}$$

where σ_θ, and σ_r are dimensionless tangential and radial stresses normalized by E_Y, respectively, providing relationships of $\sigma_r - \nu\sigma_\theta = \varepsilon_r$, and $\sigma_\theta - \nu\sigma_r = \varepsilon_\theta$, where ν is the Poisson's ratio of the thin film. Combining Eqs. S1-4 as a function of σ_r provides equations of

$$\rho\frac{d}{d\rho}\left[\frac{d}{d\rho}\left(\rho\sigma_r\right)+\sigma_r\right]+\frac{1}{2}\left(\frac{dW}{d\rho}\right)^2 = 0, \tag{S5}$$

$$\sigma_r \frac{dW}{d\rho} = -\frac{1}{2}q\rho. \tag{S6}$$

Substituting Eq. S6 in to Eq. S5 yields

$$\sigma_r^2 \frac{d}{d\rho}\left[\frac{d}{d\rho}\left(\rho\sigma_r\right)+\sigma_r\right]+\frac{1}{8}q^2 p = 0. \tag{S7}$$

The approximated solutions of Eq. S7 based on a series expansion proposed by Hencky is

$$\sigma_r\left(\rho\right) = \frac{1}{4}q^{2/3}\sum_{n=0}^{\infty} b_{2n}\rho^{2n}, \tag{S8}$$

and similarly, dimensionless out-of-plane displacement $W(\rho)$, and tangential stress $\sigma_\theta(\rho)$ can be written as

$$W(\rho)=q^{1/3}\sum_{n=0}^{\infty}a_{2n}\left(1-\rho^{2n+2}\right), \tag{S9}$$

$$\sigma_{\theta}(\rho)=\frac{1}{4}q^{2/3}\sum_{n=0}^{\infty}(2n+1)b_{2n}\rho^{2n}. \tag{S10}$$

Here, a_{2n} and b_{2n} are coefficients that depend on the Poisson's ratio of the thin film, which can be calculated using boundary conditions. The calculated a_{2n}, and b_{2n} up to n=20 based on Poisson's ratio of PDMS, ν_{PDMS}=0.5 are summarized in Table 1.

S2. Strain Calculation

We calculate the strain level of the pressurized PDMS using Hencky's model based on the measured out-of-plane displacement (see Fig. 3a in main text). As an example of the calculation, we show calculated profiles of dimensionless out-of-plane displacement $W(\rho)$, and radial $\varepsilon_r(\rho)$ and tangential strains $\varepsilon_\theta(\rho)$ based on the measured displacement at the center of the diaphragm. The center of the PDMS film is inflated with out-of-plane displacement of $u(0)$=1.2025mm when pressure level of 31.03kPa is applied to the sealed cavity. This measured displacement translates to the dimensionless out-of-plane displacement of $W(0)=w(0)/r$=1.2025mm/3.175mm=0.3787. Using Eqs. S8-10, we calculate the profile of $W(\rho)$, $\varepsilon_r(\rho)$, and $\varepsilon_\theta(\rho)$ when $W(0)$ =0.3789 (see Fig. S1). For other measured out-of-plane displacements from the PDMS bulging, we use the same calculation protocol and estimate the strain levels.

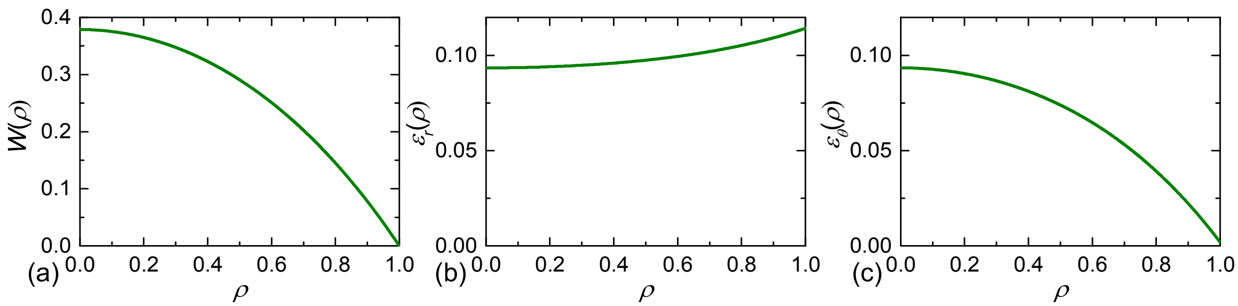

Figure S1. Calculated $W(\rho)$, $\varepsilon_r(\rho)$, and $\varepsilon_\theta(\rho)$ when the out-of-plane displacement at the center position of the PDMS film is $w(0)$=1.2025mm. (a) Calculated $W(\rho)$. (b-c) $\sigma_r(\rho)$ and $\sigma_\theta(\rho)$ are first calculated using Eqs. S8 and S10 and converted to $\varepsilon_r(\rho)$, and $\varepsilon_\theta(\rho)$ using the relationships of $\sigma_r - \nu\sigma_\theta = \varepsilon_r$, and $\sigma_\theta - \nu\sigma_r = \varepsilon_\theta$. The strain levels of $\varepsilon_r(0)$, and $\varepsilon_\theta(0)$ at the center of the PDMS film are identical $\varepsilon_r(0)=\varepsilon_\theta(0)\approx 0.0935$, proving biaxial strain.

Table S1. Calculated a_{2n} and b_{2n} for PDMS

n	0	2	4	6	8	10	12	14	16	18	20
a_n	5.421×10^{-1}	4.312×10^{-2}	7.625×10^{-3}	1.668×10^{-3}	4.055×10^{-4}	1.050×10^{-4}	2.837×10^{-5}	7.900×10^{-6}	2.251×10^{-6}	6.531×10^{-7}	1.922×10^{-7}
b_n	1.845	-2.936×10^{-1}	-3.116×10^{-2}	-5.372×10^{-3}	-1.118×10^{-3}	-2.581×10^{-4}	-6.371×10^{-5}	-1.647×10^{-5}	-4.407×10^{-6}	-1.210×10^{-6}	-3.395×10^{-7}

S3. Reversible Bulging Measurement

We perform the measurement by both increasing and decreasing the bulging at small strain levels, as shown in Figure S2. During the experiment, the PDMS with MoS_2 on top sequentially goes through zero differential pressure (Δp=0), 2.07kPa differential pressure (strain of 0.25%, at Δp=2.07kPa), back to zero pressure (Δp=0), then 4.83kPa pressure (strain of 1%, at Δp=4.83kPa), and then back to zero differential pressure again (Δp=0). PL and Raman data in Figure S2 show that after small amount of bulging, the Raman and PL signatures are close to the initial value when the differential pressure returns to zero, which proves that using bulging to tune the optical signature is a reversible process when the amount of strain on MoS_2 is small.

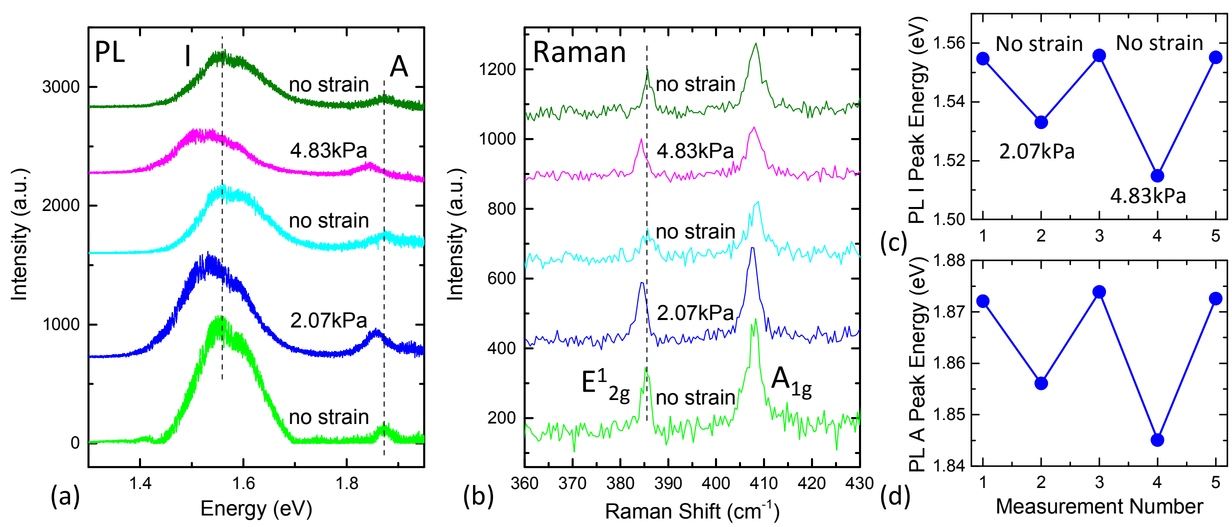

Figure S2. Reversible straining measurement of the 2L MoS_2 structure shown in Figure 1e. (a) PL spectra recorded when the PDMS goes through the process of zero differential pressure (Δp=0), 2.07kPa differential pressure (strain of 0.25%, at Δp=2.07kPa), back to zero pressure (Δp=0), under 4.83kPa pressure (strain of 1%, at Δp=4.83kPa), and then back to zero differential pressure again (Δp=0). (b) Raman spectra recorded during the same bulging process. (c) & (d)

Summary of (c) I peak and (d) A peak energy during the measurement, showing that the peak positions return very close to the initial position under small strain.

Reference

[1] Fichter, W. *NASA Technical Paper* **1997**, *3658*.