Supporting Information

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Engineered Creation of Periodic Giant, Nonuniform Strains in MoS₂ Monolayers

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Supporting Note 1: Engineering the dome formation process.

In Figure 1 of this Note we show the sketch of a typical pattern designed for our samples. The pattern is realized by depositing a film with uniform thickness of hydrogen silesquioxane (HSQ) on the sample. A negative tone e-beam resist is used, so that the desired pattern (wine areas in the figure) can be obtained by electron-irradiating the whole sample's surface apart for the white areas visible in the figure (octagonal openings and corridors) where the domes will form upon proton-irradiation.

![Figure 1 of Note 1. Design and realization of the HSQ masks.](image)

Sketch of a typical mask designed for patterning the samples: arrays of openings (in white) with different characteristics are arranged in different squares (in wine) with same dimensions. S indicates the diameter of the openings. The sketch here shown might represent a basic module to be repeated periodically all over the sample.

The choice of realizing octagonal openings rather than circular ones is aimed at reducing the electron beam lithography exposure time while still creating openings with a circular-like shape. After the electron exposure, the resist is developed in an aqueous solution of tetramethyl ammonium hydroxide at 2.4% to remove the unexposed resist and leave the desired openings (see Experimental Section in the main text). With this procedure, several arrays with openings (white areas) of different sizes are arranged in different squares (in wine) with same dimensions. The size of the openings, their number and disposition, and the distance between the squares can be varied according to the specific needs. Once the basic ingredients are chosen, the same module (like the one represented in Figure 1) is repeated periodically all over the sample surface where the flakes had been previously deposited.

This approach was first presented in our previous work Ref. 1. In that case, this method allowed to control the nucleation sites of the domes and to achieve a narrower size distribution, while the degree of homogeneity of the sample was still non optimum, as shown in Figure 2 of this Note. In addition, in those patterned domes the aspect ratio -and hence the strain field- does not differ from that found in random domes,
Figure 2 of Note 1. Previous attempt to realize patterned samples.
AFM image of the patterned WS2 sample realized in Ref. 1. Array with size $S = 1 \, \mu m$ (A) and $S = 5 \, \mu m$ (B), showing how the domes were not entirely filling the openings.

To optimize irradiation, we here realized a sample with a 30-mn-thick mask and we first irradiated it with dose $d_0 = 5.5 \cdot 10^{16}$ protons/cm$^2$ (higher than in Ref. 1, where it was equal to $4.0 \cdot 10^{16}$ protons/cm$^2$) as schematized in Figure 3A of this Note. This first treatment resulted in the formation of domes still not filling the openings (see AFM image shown as inset). The sample was then re-irradiated with dose $d_1 = 2.0 \cdot 10^{16}$ protons/cm$^2$, and the domes’ footprint was brought to nearly totally occupy the openings (see panel B). A comparison between the filling percentages after the first and second irradiation is shown in panel C, where we provide also a comparison with the sample of Ref. 1. Following this result, all the samples prepared for this work were irradiated with doses equal to $6-7 \cdot 10^{16}$ protons/cm$^2$, representing the best trade-off between high filling and high formation yield (for too high doses, the domes start exploding).
**Figure 3 of Note 1. Optimization of the proton-irradiation process.**

(A) Sketch of a patterned (wine area) sample (gray part) irradiated with protons (orange particles) with dose $d_0$. The inset is an AFM 3D image of a typical dome (in an opening with diameter $S = 5 \, \mu m$) formed in a sample patterned with a 30-nm-thick mask and irradiated with proton dose $d_0 = 5.5 \times 10^{16}$ protons/cm$^2$. The dome is not totally filling the opening, as demonstrated by the presence of the black region around the dome footprint. (B) Same as panel a, but for the use of a higher proton dose $d_0 + d_1$, with $d_1 = 2.0 \times 10^{16}$ protons/cm$^2$. In this case, a typical dome (in an opening with diameter $S = 5 \, \mu m$) almost entirely occupies the opening. (C) Histograms of the filling percentages (footprint of the dome divided by the area of the opening) for the sample described in panel A and B after irradiation with proton dose $d_0$ (blue columns) and $d_0 + d_1$ (orange columns), for different opening sizes. The filling percentages obtained in Ref. 1 are also shown (pink columns) for comparison.
Supporting Note 2: Developing a strain engineering protocol.

Here we provide more details about the realization of highly constrained domes via increasing the resist thickness.

First, we here discuss the numerical simulations performed with Comsol to simulate the effect of a constraint in the dome formation process. In Figure 1A of this Note we show the profile simulated for a typical dome (black line) with aspect ratio $h_m/R = 0.16$ (where $R = 500$ nm). To study the role of a boundary, we take a dome with a radius value increased by 10 % and mimic the effect of the resist via introducing external forces and an extra-pressure applied to the membrane, until matching the $R$ value of the typical dome (see red line in panel A). This leads to an aspect ratio of 0.26, and to a remarkable increase of both the in-plane strain components (i.e., the radial component shown in panel B and the circumferential component shown in panel C).

Figure 1 of Note 2. Simulating the effect of a constraint. (A) Numerical simulations of the height profiles of a typical dome with universal aspect ratio (black line) and of a dome subjected to external forces (red line). These latter are aimed at mimicking the role played by a strong enough resist, acting as a constraint to the 2D membrane. The inset shows the 3D AFM image of a real dome and depicts the meaning of maximum height and footprint radius. (B) Radial strain component (whose meaning is sketched in the inset) simulated for the two cases in panel A. (C) Same for the circumferential strain component (whose meaning is sketched in the inset).
To show the real effect played by a boundary, in Figure 2 of this Note we present a comparison between the optical images of two different samples. The optical image of a MoS$_2$ sample patterned with a 30-nm-thick mask -after dome formation- is shown in Figure 2A. Indeed, the domes which formed within the openings have typically a regular size, while the presence of domes with random size can be observed in the corridors between the squares. In panel B, we show the optical image of another patterned MoS$_2$ sample. In this case the thickness of the mask is about 70 nm. The domes within the openings are almost totally filling the openings and, in this case, the thick mask represents a constraint for the domes. This is particularly evident by looking at the corridors between the different squares, where elliptically-deformed domes formed, such as those within the cyan rectangle. Notice that the formation of elliptically-deformed domes differ sizeably from the phenomenology shown in the main text, Figure 1d: In that case the dome was able to raise a 50-nm-thick resist and was not deformed due to its presence.

Figure 2 of Note 2. Design and realization of the HSQ masks.

(A) Optical image of a MoS$_2$ sample after patterning (where the thickness of the mask, $t$, is about 30 nm) and dome formation. Here the opening sizes in the four squares are 3 µm (on a 6 × 6 array, top left), 5 µm (on a 5 × 5 array, top right), 1 µm (on a 11 × 11 array, bottom left) and 3 µm (on a 6 × 6 array, bottom right). Indeed, many domes with random sizes formed within the corridors that separate the squares. (B) Optical image of another MoS$_2$ sample after patterning (with a 70-nm-thick mask) and dome formation. Here the opening sizes in the two squares are 1 µm (on a 11 × 11 array, top) and 3 µm (on a 6 × 6 array, bottom). In this case, many domes in the corridors (see cyan rectangle) are elliptical due to the anisotropic constraint due to the thick mask around.

Masks with different thicknesses were realized for the studies presented in this work. While the height of the mask is not perfectly constant all over the sample –due to the small sizes the of samples that
are typically processed and to the presence of the flakes on the surface—with our method we are able to realize masks of a certain height within about 10 nm. In any event, the heights of the masks were all measured by AFM, flake by flake (as exemplified in Figure 3 of this Note), so that the thickness values quoted in this work were all determined experimentally. Our studies show that for masks with height between 50 and 100 nm, we can achieve high aspect ratios of the domes, still having a dome formation process characterized by a high yield, > 95 % for medium and small domes and ~ 60-70 % for big domes, as one can see in panels A and B of Figure 2. The yield of the formation process is indeed also influenced by the proton-dose. For low doses, the domes form almost within all the openings. If the dose is brought to an optimum level, the domes almost totally fill the openings, as discussed in Supporting Note 1. However, if higher doses are used, the domes start exploding, as a consequence of the too high internal pressure exerted on the membrane by the trapped hydrogen, and of the boundary role played by the mask.

Figure 3 of Note 2. Design and realization of the HSQ masks.
(A-B) AFM images of two corridors in two different samples patterned with masks with different thicknesses. (C) AFM profile across the corridor in panel A, showing that the thickness is about 45 nm. (D) Same for the corridor in panel B: here the thickness is higher than 50 nm.
Supporting Note 3: Patterned domes and strain fields.

As discussed in Refs. 1 and 2, finite element method (FEM) calculations can be used to model the height profile and strain tensor -in spherical coordinates- of the domes. This allows us to estimate how the strain tensor varies across the dome surface, while keeping into account the elastic properties of the 2D MoS$_2$ membrane (the same elastic tensor of ref. 1 has been used in this work). As shown in Supporting Note 2 and as expected from theoretical models$^3$, the presence of external constraints - that lead to an increased aspect ratio - should lead to an increase of both the in-plane strain components, while their analytical dependence on the radial coordinate should not be modified. While in Supporting Note 2 arbitrary forces had been included to simulate the presence of the mask, here we avoid introducing arbitrary forces by totally clamping the 2D membrane at its edges. This procedure is justified by the experimental observation that -in the case of thick resists- the dome is not able to raise the mask and its dimensions are thus determined by the size of the opening. For the numerical calculations, we thus clamp the membrane edges by fixing the value of the radius so to match the experimental value, imposed by the opening size. We then apply an internal upward pressure to simulate the presence of the pressurized molecular hydrogen within the dome. The pressure is increased until the height of the simulated dome corresponds to the experimental height value. As shown in Fig. 3 of the main text, this procedure allows to well reproduce the experimental profile of the domes. In the case of the random domes, the membrane is clamped as well at the edges to simulate the role played by the adhesion forces in determining the dome’s radius. The upward pressure applied in this case aims at mimicking the presence of the gas at equilibrium conditions$^3$ in the absence of external factors, which is achieved again by matching the equilibrium height measured by AFM. The pressure values obtained for random domes are thus consistent with previous estimations for random bubbles in other systems$^4$, while remarkably higher pressures (up to an order of magnitude) are needed for the engineered domes. In Figure 1A of this Note (upper panel) we show the simulated strain tensor components as a function of the distance from the dome centre, for the same random dome of Fig. 3 of the main text (with footprint radius $R = 2200$ nm and maximum height $h_m = 363$ nm, resulting in $h_m/R = 0.165$). In the lower panel we calculate the degree of isotropy of the in-plane strain acting on the dome’s surface, defined as:

$$\text{Strain Isotropy} = 1 - \frac{\varepsilon_r - \varepsilon_t}{\varepsilon_r + \varepsilon_t}.$$
Figure 1 of Note 3. Strain distribution in patterned domes.

(A) Upper panel: Strain components in polar coordinates (\(\varepsilon_r\) = radial component, \(\varepsilon_t\) = circumferential component, \(\varepsilon_z\) = perpendicular component\(^1\)) calculated for the same random dome of Fig. 3 of the main text. The components were calculated via FEM calculations as a function of the distance from the centre of the dome, \(|r|\). Lower panel: Degree of isotropy of the in-plane strain acting on the dome. (B-D) Same for the three patterned domes of Fig. 3 of the main text. The three domes have: \(R = 2343\) nm and \(h_m = 436\) nm, resulting in \(h_m/R = 0.186\) (B); \(R = 1414\) nm and \(h_m = 310\) nm, resulting in \(h_m/R = 0.219\) (C); \(R = 515.5\) nm and \(h_m = 136.1\) nm, resulting in \(h_m/R = 0.264\) (D).

While the sum of the two in-plane components (i.e., the radial, \(\varepsilon_r\), and circumferential, \(\varepsilon_t\), components\(^1\)) is the relevant quantity to be considered for the strain-induced variation of the electronic properties of TMDs, the degree of isotropy should be taken into account when considering the vibrational properties of the strained membrane and its intertwined spin/valley degrees of freedom. As shown here, according to our calculations strain is isotropic at the center of the domes, while it becomes uniaxial - and therefore totally anisotropic - at the edges. The real behavior at the edges might not be perfectly described by our model, due to both the transition from bulk to dome, the boundary role played by the mask in patterned domes, and the presence of small domes at the edges of the large domes (typically observed mostly in random domes), that might lead to deviations from the ideal situations described by numerical simulations\(^2\). In any event, one should expect strain to be strongly anisotropic at the edges. The same calculations of panel A are repeated for the three patterned domes of Fig. 3 of the main text in panels b-d, showing that the same qualitative behaviour holds for both the patterned and the random domes. Quantitatively, there is instead a remarkable difference, very high strain values being achieved for small patterned domes, close to the rupture critical value, according to previous studies\(^5,6\). Such a result is of great interest for both fundamental
and applicative prospects. The creation of high and strongly anisotropic strains can give rise to exciton anisotropy splittings of interest for valley- and spintronics\textsuperscript{7}. The possibility to generate strain gradients over micrometric region is potentially interesting for applications in the fields of photovoltaics, photocatalysis and photodetection\textsuperscript{8}. In particular, strain has been demonstrated to change the band structure of TMDs in such a way that the exciton energy shifts by several tens of meV/%\textsuperscript{2,6,9}. Additionally, in presence of a seamless strain variation, the band gap reduction for increasing strain induces funnelling phenomena, where the excitons drift for hundreds of nm before recombining\textsuperscript{2,8,10}. In addition, it has been recently demonstrated that strains between 1 and 4 % in WS\textsubscript{2}, MoS\textsubscript{2} and WSe\textsubscript{2} turn the band gap from direct to indirect, with indirect excitons characterized by much longer decay times\textsuperscript{2}. The generation of periodic and high strain gradients allow therefore at the same time to absorb energy over a relatively large portion of the electromagnetic spectrum and to harvest the photogenerated carriers in particular position, which is particularly promising for the realization of efficient solar cells\textsuperscript{8}.

As for the possibility to absorb light, with patterned arrays of medium domes, for instance, we get a variable total in-plane strain between 3 and 7 %. According to the shift rates for the excitons measured in Ref. 2, this would allow to absorb energy in a wide wavelength range, thus covering a significant portion of the solar spectrum, as shown in Figure 2 of this Note.

![Figure 2 of Note 3. Absorption of a wide portion of the solar spectrum on earth by patterned domes.](image-url)

The yellow area represents the solar spectrum on earth according to the active standard ASTM G173 - 03(2012). The red area represents approximately the portion of the spectrum that would be absorbed by medium patterned domes, according to the shift rates estimated for the direct and indirect excitons in Ref. 2.
In addition to the great potentiality of our system for photovoltaics applications, the possibility to generate strain fields in controllable positions holds great relevance also because it paves the way for the observation of novel physical phenomena. In particular, strain fields in graphene have been predicted and demonstrated to give rise to pseudomagnetic fields, up to hundreds of Tesla\textsuperscript{11,12,13,14}. Additionally, periodic configurations of magnetic fields in 2D electron gases have been predicted to allow current to flow without dissipation\textsuperscript{15}, which is similar to superconductivity. While the insurgence of pseudomagnetic fields in strained graphene has been assessed as a manifestation of the chirality of the Bloch wave functions around the two inequivalent corners of the hexagonal Brillouin zone, TMD MLs combine these effects with a strong spin-orbit coupling (SOC)\textsuperscript{16}. As a consequence, it has been predicted that in mechanically deformed TMD MLs the large spin-orbit coupling rotates the wave function in the spinor basis, and a gauge field may arise\textsuperscript{17}. 
Supporting Note 4: Analysis of the Raman shifts in random and patterned domes.

Here we discuss micro-Raman (μ-Raman) measurements aimed at confirming that our domes are subjected to high strains as a consequence of the high aspect ratios measured by AFM. As a general fact, strain has been demonstrated to lead to a softening of the Raman modes\(^1,2,6,9,18,19,20\). The quantitative effect of strain on the Raman modes, however, is dependent on the kind of strain that is applied (i.e., its orientation and anisotropy). For this reason, we performed our measurements in the point of maximum symmetry for our domes, that is, the centre. There, strain is known to be isotropic biaxial and a simple correspondence with the aspect ratio has been established by models developed in the framework of the membrane theory\(^1,2,6,21,22\):

\[
\varepsilon_p = 2 \cdot f(\nu) \cdot \left(\frac{h_m}{R}\right)^2,
\]

with \(f(\nu) = 0.721\) for MoS\(_2\) domes, where \(\varepsilon_p\) is the total in-plane strain. As for the patterned domes, in the case of the small domes (opening with size \(S = 1 \mu m\)) we were limited by the resolution of our optical setup (the laser spot has dimensions comparable to the opening, see Experimental Section in the main text and Figure 5a of the main text). For big domes, we would have had the best resolution, but the aspect ratio is only marginally larger than in random domes. For this reason, medium domes in openings with \(S = 3 \mu m\) represented the best trade-off, having significantly higher aspect ratios and allowing to be measured with relatively good spatial resolution. As for random domes, in order to have a comparison on an equal footing, domes with diameter of about 3 \(\mu m\) were chosen. The results obtained for the in-plane \(E_{2g}^1\) mode have been discussed in the main text. The redshifts measured for the out-of-plane \(A_{1g}\) Raman peak (see, e.g., spectra in Fig. 5c of the main text) with respect to the strain-free ML at the summit of random domes (purple) and patterned domes (green) are instead displayed in Figure 1 of this Note.

![Figure 1 of Note 4. Redshift of the A\(_{1g}\) Raman peak in random and patterned domes.](image)

Redshifts measured for the \(A_{1g}\) Raman peak (see, e.g., spectra in Fig. 5c of the main text) with respect to the strain-free ML at the summit of patterned (openings with size \(S = 3 \mu m\)) domes (green) and of random domes with similar dimensions (purple). The data are plotted as a function of...
the total in-plain strain at the summit, $\varepsilon_{p}^{\text{summit}}$, analogously to Fig. 5d of the main text. The grey line is a linear fit to the data.

The grey line is a linear fit to the data, showing that the data do not follow the expected linear behaviour with strain. A deviation from linearity has been observed also in WS$_2$ domes in Ref 18, and can be ascribed to the strong intensity modulation of this mode with strain, as shown and discussed in Refs. 6 and 18. In particular, notice that according to Ref. 6 the intensity of the $A_{1g}$ mode exhibits a maximum for a biaxial strain $\sim 3\%$ (corresponding to $\varepsilon_{p}^{\text{summit}} \sim 6\%$) and a minimum for a biaxial strain $\sim 2\%$ (corresponding to $\varepsilon_{p}^{\text{summit}} \sim 4\%$), that approximately correspond to the strain values at the top of patterned and random domes, respectively. This effect, coupled to the limited resolution of our setup compared to the dimensions of the domes, might cause the data concerning random domes to deviate from the expected linear behaviour.

Figure 2 of Note 4. Raman mapping along a radius of a large random dome. (A) Colormap corresponding to a µ-Raman scan across a random MoS$_2$ dome while moving from an edge towards the centre. A relatively large dome with radius 2.35 µm was chosen in order to achieve a good spatial resolution. The horizontal axis indicates the laser spot position with respect to the dome centre ($r$), whereas the vertical axis indicates the Raman shift with respect to the laser line (laser wavelength = 532.2 nm, see Experimental Section in the main text). The µ-Raman
intensity is shown in a false colour scale (see colour bar). The mode at 408.6 cm$^{-1}$ is the A$_{1g}$ mode of the bulk flake beneath the dome. The A$_{1g}$ mode of the dome clearly features both a redshift, while moving from the edge towards the centre, and a strong modulation of intensity, which is ascribable to the strain variation along the scanning direction$^6$. (B) Integrated intensity of the A$_{1g}$ mode for the dome shown in panel B. The pink and blue lines highlight the positions of the edge and the centre of the dome, respectively.

To verify this hypothesis, we acquired the Raman spectra at the summit of random domes with larger diameter, that allows to get better resolved measurements. In Figure 2A of this Note we show a µ-Raman scan across a random MoS$_2$ dome while moving from an edge towards the centre, for a dome with radius 2.35 µm and $h_m/R = 0.164$. The faint horizontal signal at 408.6 cm$^{-1}$ is the A$_{1g}$ mode of the bulk flake beneath the dome. This mode features a tiny variation of intensity, ascribable to dome-induced interferential phenomena, and remains constant in energy. The A$_{1g}$ mode of the dome, on the contrary, clearly features both a redshift (while moving from the edge towards the centre) and a strong modulation of intensity, which is therefore chiefly ascribable to the strain variation along the scanning direction$^6$. To better appreciate the intensity variation while going from the edge to the centre, in panel B we display the integrated intensity as a function of the radial coordinate, $r$. Indeed, the maximum intensity is reached close to the edge, where the shift with respect to the unstrained ML is 0.7 cm$^{-1}$. The intensity drops at a distance of about 1 µm from the centre, to re-increase a little bit towards the centre, where the Raman shift equals 2.9 cm$^{-1}$. Notice that for the smaller random domes in Figure 1 of this Note the shift measured at their summit is also equal to 0.7 cm$^{-1}$. This value is the same found towards the edge of the larger dome, where the intensity is maximum. As a matter of fact, when measuring smaller domes, the Raman spectrum is the result of a convolution within the exciting laser spot (gaussian with $\sigma = 0.23$ µm, see Experimental Section in the main text), and the component with maximum intensity could dominate. Furthermore, the relative intensity of the two maxima in Figure 2B could be different in smaller domes (due to the different role played by interference$^{18}$) and further favour the edge component. This is possibly the reason for the deviation from the expected behaviour for this mode, and the different shift measured for random domes with analogous aspect ratio but different radii (shift ~ 0.7 cm$^{-1}$ in domes with diameter ~ 3 µm, as shown in Figure 1, and 2.9 cm$^{-1}$ in the dome with diameter ~ 4.7 µm discussed in Figure 2). The in-plane E$_{2g}^1$ mode could be as well affected by a similar phenomenology, but the intensity of this mode with strain has been shown to be less subjected to oscillating behaviours for the strain values considered$^6$.

In order to have a quantitative feedback of the above discussion, in Table 1 of this Note we report the redshift rates and Grüneisen parameters for the E$_{2g}^1$ and A$_{1g}$ Raman modes, calculated by
considering only random domes, only patterned domes, and all the domes. For the reasons discussed above, a large difference in the redshift rate - and consequently in the Grüneisen parameter - is found for the $A_{1g}$ mode by considering the different sets of data, where the data concerning random domes are affected by the oscillating behaviour of this mode with strain. On the contrary, the redshift rate of the $E_{2g}^1$ mode varies little depending on the set of data considered, confirming that this mode is less subjected to oscillations in the intensity for increasing strain. Notice also that the values found for the $E_{2g}^1$ mode are in agreement with those reported in previous works, as discussed in the main text. In addition, the Grüneisen parameter found for the $A_{1g}$ mode in patterned domes agrees well with previous results: $\gamma_{A_{1g}} = 0.21$ in Ref. 6 where strain is biaxial and $\gamma_{A_{1g}} = 0.21$ in ref. 23 where strain is uniaxial.

|                  | Only random | Only patterned | All data |
|------------------|-------------|----------------|----------|
| $\Delta_{E_{2g}^1}$ (cm$^{-1}$/%) | 2.40±0.05   | 2.15±0.05   | 2.2±0.1  |
| $\gamma_{E_{2g}^1}$            | 0.62±0.02   | 0.56±0.02   | 0.58±0.03|
| $\Delta_{A_{1g}}$ (cm$^{-1}$/%)  | 0.30±0.05   | 0.80±0.05   | 0.65±0.10|
| $\gamma_{A_{1g}}$              | 0.07±0.01   | 0.20±0.01   | 0.17±0.03|

Table 1 of Note 4. Redshift rates and Grüneisen parameters.

Redshift rates for in-plane strain ($\Delta$) and Grüneisen parameters ($\gamma$) for the in-plane $E_{2g}^1$ and out-of-plane $A_{1g}$ Raman modes in MoS$_2$ monolayers under isotropic strain (domes’ summit). The displayed values were calculated from the data in Fig. 5d of the main text and Figure 1 of this Note, by considering only the set of data concerning random domes, only that concerning patterned domes, and both the two sets.
**Supporting Figure S1: Probing the thickness of the domes via second harmonic generation.**

(A) Intensity of the second harmonic (SH) signal (navy points) during a scan performed on a patterned sample ($S = 3 \, \mu$m). The measurements were performed by exciting at 900 nm and collecting the signal at 450 nm (see Experimental Section in the main text). The scan was performed by moving along a given direction at steps of 250 nm, in such a way to acquire the measurements along the diameter of two domes. The correspondence between the laser spot position and the domes is established by panel B. As here shown, second harmonic generation (SHG) is obtained only when scanning the laser over the two domes. The solid line is a fit to the intensity data with gaussian functions. Indeed, MoS$_2$ is characterized by the ability to originate bright SHG in the monolayer limit, while the SH signal decreases rapidly as the number of layers increases$^{24,25}$. In particular, even number of layers do not give rise to SHG, while the SH of the tri-layer is about 5 times lower than that of the ML. SHG measurements can be therefore exploited to estimate the number of layers and were thus performed on several domes, as for the two domes shown here. The intensity of the SH signal was found to be always comparable or larger than in the ML for all the measured domes, thus suggesting a single-layer thickness of the domes, as schematized in the inset. (B) 3D AFM image of the two domes on which SHG measurements were performed.
Supporting Figure S2: Indentation of a dome by the AFM tip.

(A) Sketch of the indentation experiment by the AFM tip. A tip with curvature radius equal to 40 nm is positioned at the centre of the dome (same dome of Figure 4c, left, in the main text) and pushed down until reaching the substrate. (B) AFM profile along a diameter of the dome before indentation (black points) and after the indentation experiment (red points, corresponding to the AFM image of Figure 4c, right panel). The height profiles have been normalized to the footprint radii of the dome to highlight the different aspect ratios.
Supporting Figure S3: Micro-photoluminescence on patterned domes.

Micro-photoluminescence (μ-PL, see Experimental Section in the main text) spectra acquired at the summit of patterned and random domes. The spectrum concerning the random dome (bottom, purple) is the same of Ref. 2 and is plotted here for comparison. This spectrum was acquired on a dome with $h_{m}/R = 0.164$ (corresponding to $\varepsilon_{p}^{\text{summit}} = 3.9\%$). While the unstrained MoS$_{2}$ ML is characterized by a direct gap (involving the K point of both conduction band, CB, and valence band, VB), strains higher than ~1.8 % turn the band gap from direct to indirect\(^2\) (the maximum of the VB turns from the K point to the Γ point\(^{26}\)). Here, the strain is higher than the crossover value, and both the direct (A, corresponding to the K$_{CB}$-K$_{VB}$ transition) and indirect (I, corresponding to the K$_{CB}$-Γ$_{VB}$ transition) exciton transitions can be observed. The purple solid lines highlight the position of the indirect and direct excitons for the random dome. In the middle, we show the spectrum (in red) of a dome created within openings with size $S = 5\ \mu$m. The dome has $h_{m}/R = 0.180$ ($\varepsilon_{p}^{\text{summit}} = 4.7\%$), and an evident strain-induced redshift of both the exciton transitions can be observed. The red solid lines highlight the position of the exciton peaks, as a guideline for the eye. Finally, the green spectrum on top was acquired on a dome created in an opening with size $S = 3\ \mu$m, and is characterized by $h_{m}/R = 0.213$ ($\varepsilon_{p}^{\text{summit}} = 6.5\%$). In this case, the exciton transitions are further redshifted, with a lower shift-rate that is due to the non-optimum resolution of our optical setup compared to the dimension of the dome (see Fig. 4a in the main text and discussion in Supporting Note 3).
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