Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Optical Grade Transformation of Monolayer Transition Metal Dichalcogenides via Encapsulation Annealing

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Defect density calculation

	Total Se atomic positions (N_{pse})	Expected Se atom number ($2^* N_{pse}$)	Total V_{Se}	Total V_{Se2}	Defect density (%) $\left(\frac{V_{Se} + 2 \times V_{Se2}}{2 \times N_{pse}}\right)$
As-stacked	62605	125210	409	2	0.33
Annealed	69257	138514	1901	3	1.38

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By utilizing ResUNet deep learning model, we extracted the total atomic positions of Selenium, expected Selenium atom number, total selenium vacancies, total diselenium vacancies. Through the extracted data the defect density was calculated using the following equation.

$$\text{Defect density (\%)} = \left(\frac{V_{Se} + 2 \times V_{Se2}}{2 \times N_{pse}}\right)$$

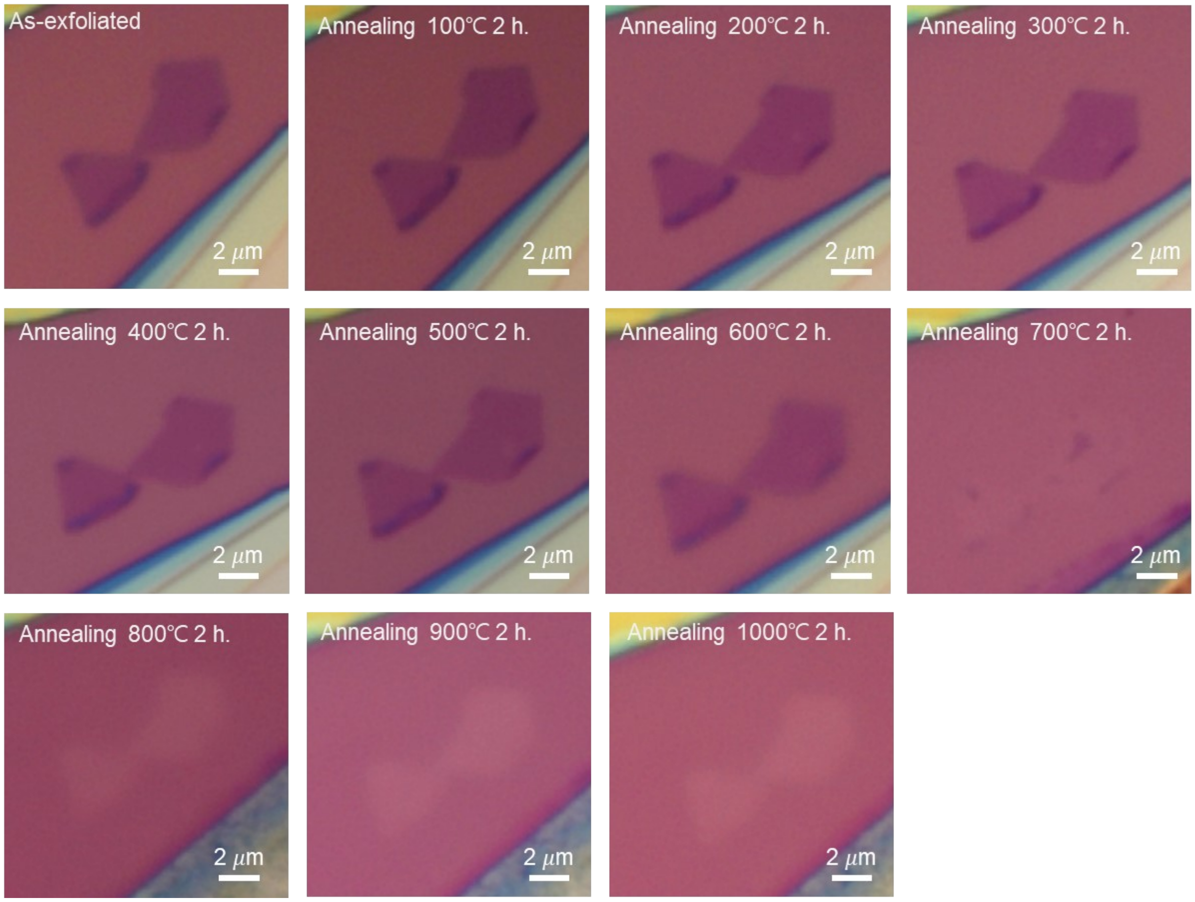

Figure S1. Optical microscopic images of exfoliated 1L-WSe_2 before and after annealing at 100-1000°C for 2h. In contrast to hBN-encapsulated 1L-WSe_2, there was a significant difference in the exfoliated 1L-WSe_2 indicating thermal decomposition.

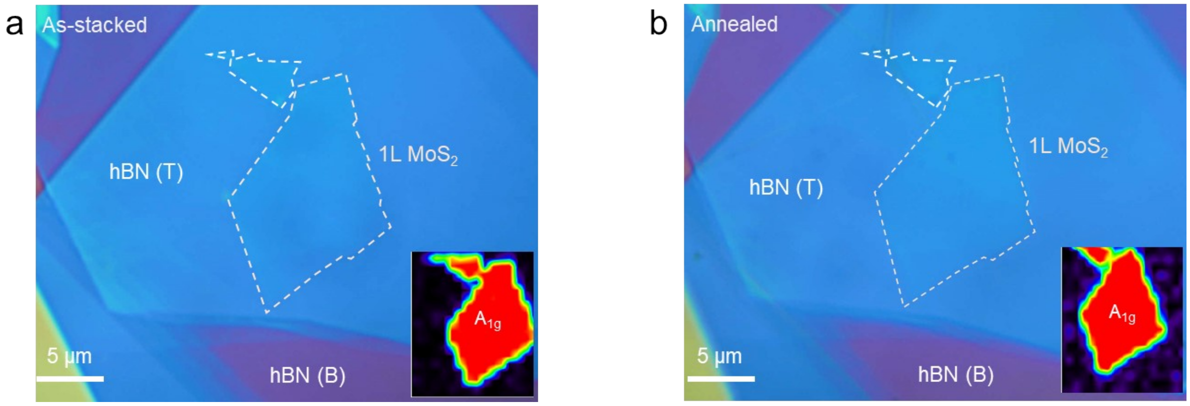

Figure S2. Optical microscopic images of hBN-encapsulated 1L-MoS_2 (a) before and (b) after annealing at 900℃ for 3 h. Inset Raman peak intensity maps for the A_{1g} mode of MoS_2.

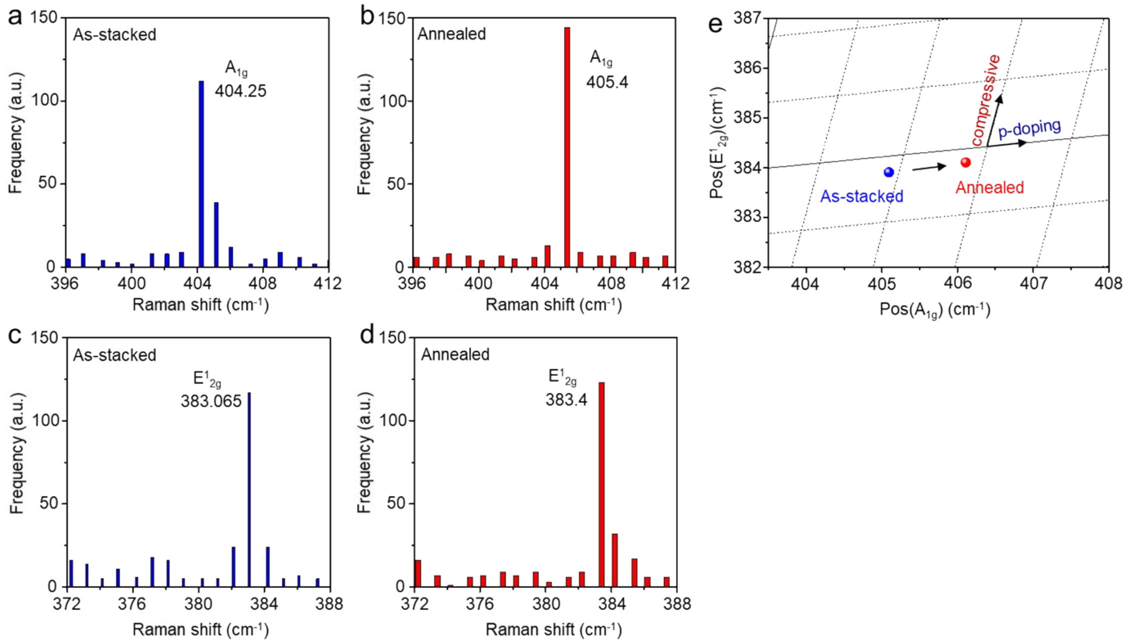

Figure S3. Raman peak shift of hBN-encapsulated 1L-MoS_2 before and after annealing. Histogram of (a,b) A_{1g} peak position, (c,d) E^1_{2g} peak position of MoS_2 before and after annealing at 900°C for 3 h. (e) A_{1g} and E^1_{2g} peak position plots for hBN-encapsulated 1L-MoS_2 before and after encapsulation annealing. A_{1g} peak and E^1_{2g} peak position was plotted based on the peak position with the highest frequency in the histogram. The shift in the peak position indicates that the MoS_2 is dedoped by encapsulation annealing.

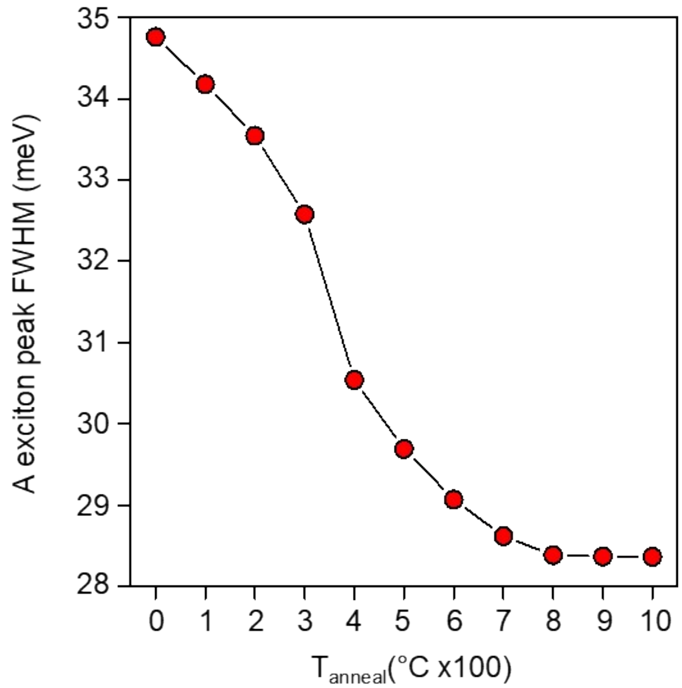

Figure S4. Annealing temperature dependent A exciton peak FWHM of hBN-encapsulated 1L-WSe_2. The FWHM decreases as the annealing temperature increases until 800°C.

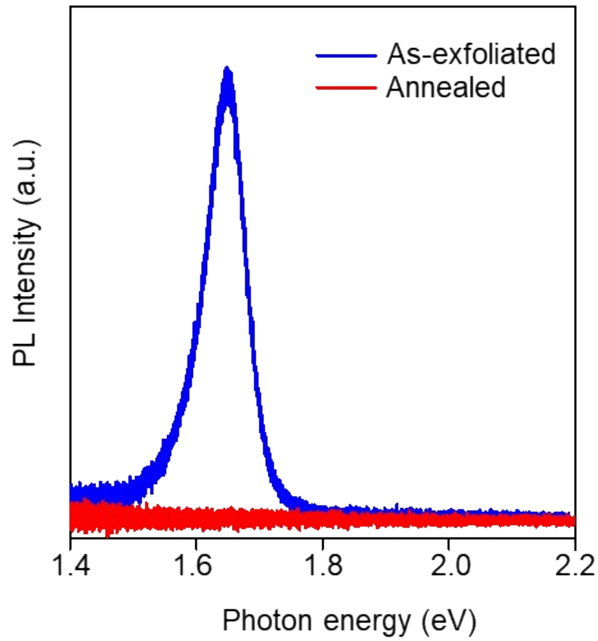

Figure S5. PL Intensity spectra of as-exfoliated 1L-WSe_2 before and after annealing at 1000°C for 2h. The PL was quenched after annealing which indicates thermal decomposition of the 1L-WSe_2.

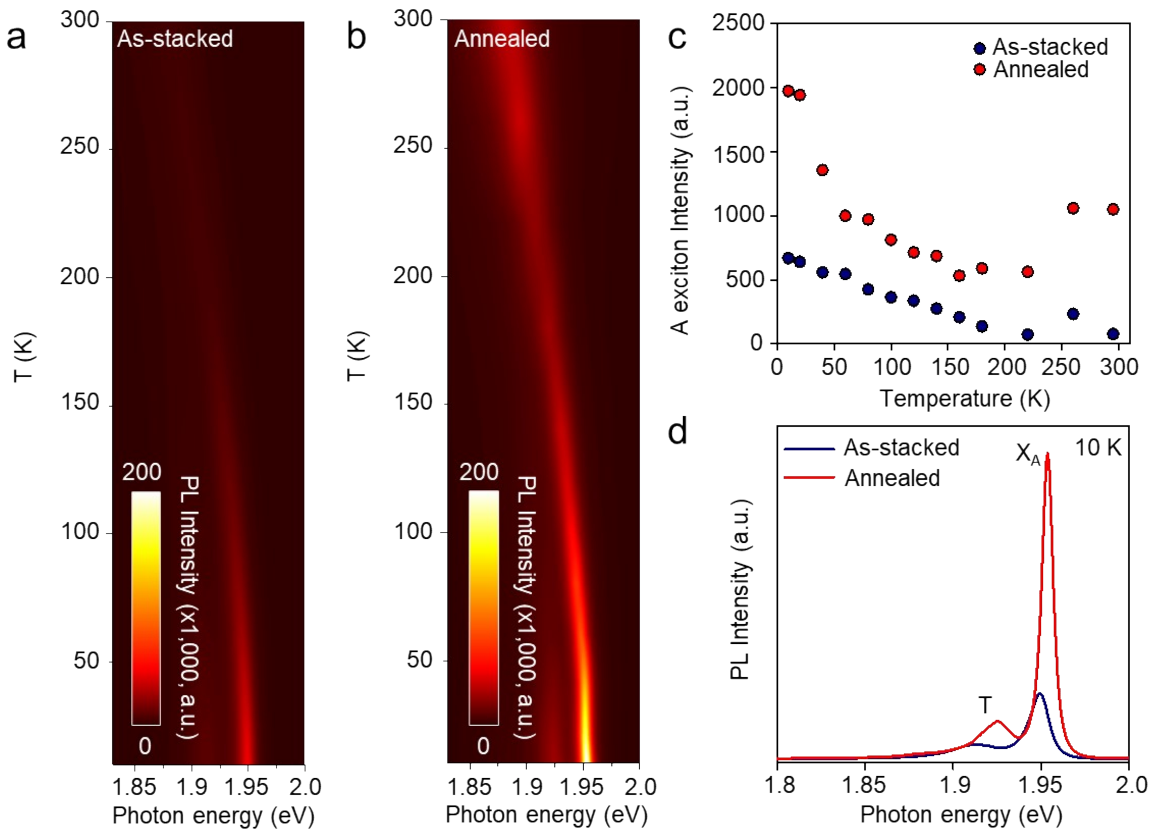

Figure S6. Contour plots of PL spectra of 1L-MoS_2 as a function of temperature (from 10 to 300K) (a) before and (b) after annealing. (c) A exciton intensity as a function of temperature of hBN-encapsulated 1L-MoS_2 before (blue) and after annealing (red). (d) PL spectra (10K) of hBN-encapsulated 1L-MoS_2 before (blue) and after annealing (red). The PL intensity of the 1L-MoS_2 increases as the temperature decreases due dominant population of bright excitons at low temperature. The PL intensity of the annealed 1L-MoS_2 was higher than that of as-stacked 1L-MoS_2 across the entire temperature range from 10 to 300K.

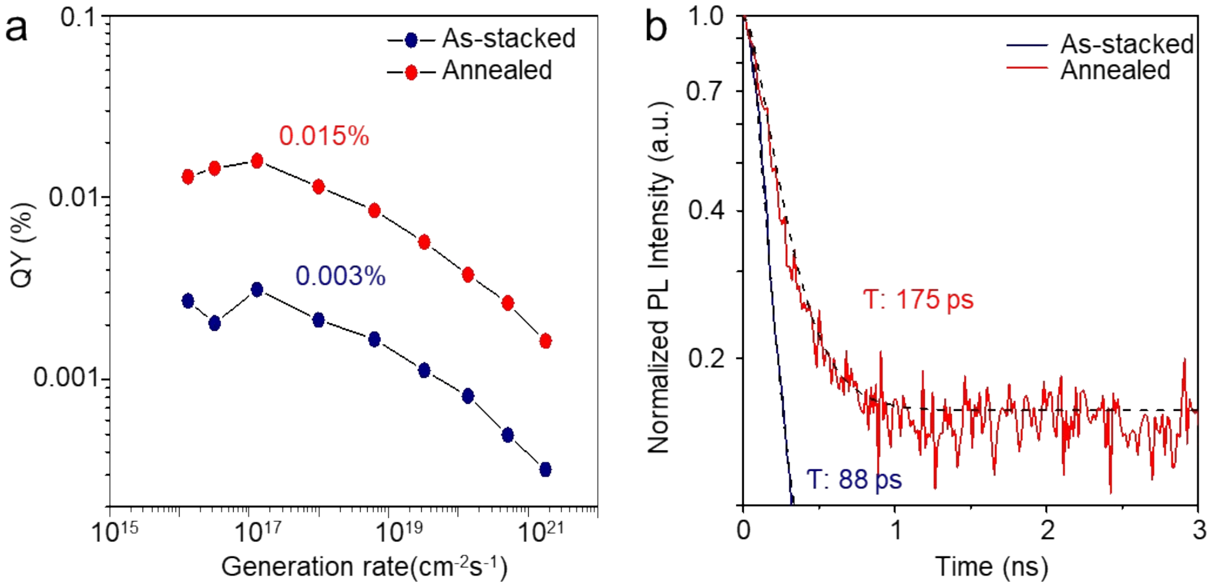

Figure S7. (a) Plot of PL QYs as a function of the generation rate and (b) normalized TRPL decay curves for hBN-encapsulated 1L-MoS_2 before (blue) and after annealing (red).-