

Supporting Information for

Effects of Post-Transfer Annealing and Substrate Interactions on the Photoluminescence of 2D/3D Monolayer WS₂/Ge Heterostructures

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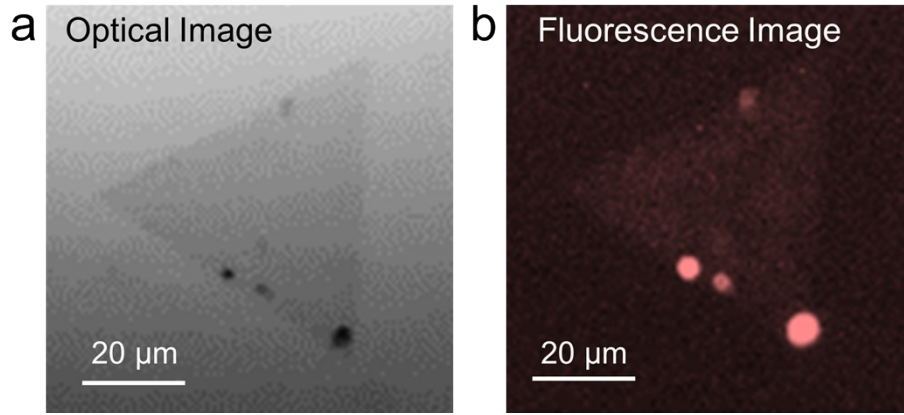
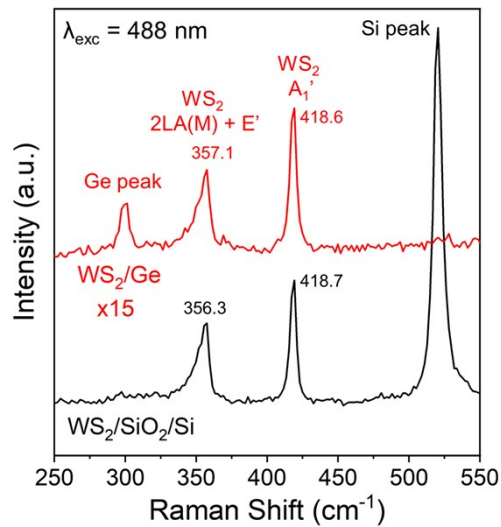


Figure S1. Typical (a) optical and (b) fluorescence images of monolayer WS₂/Ge heterostructures. A weak fluorescence emission intensity is observed in WS₂/Ge



heterostructures.

Figure S2. Raman spectra of as-grown WS₂ on SiO₂/Si and WS₂/Ge (annealed) obtained with a 488 nm laser. Note that WS₂/Ge heterostructures display a much weaker Raman signal, and the corresponding Raman spectrum data (red) is multiplied by 15 for visualization.

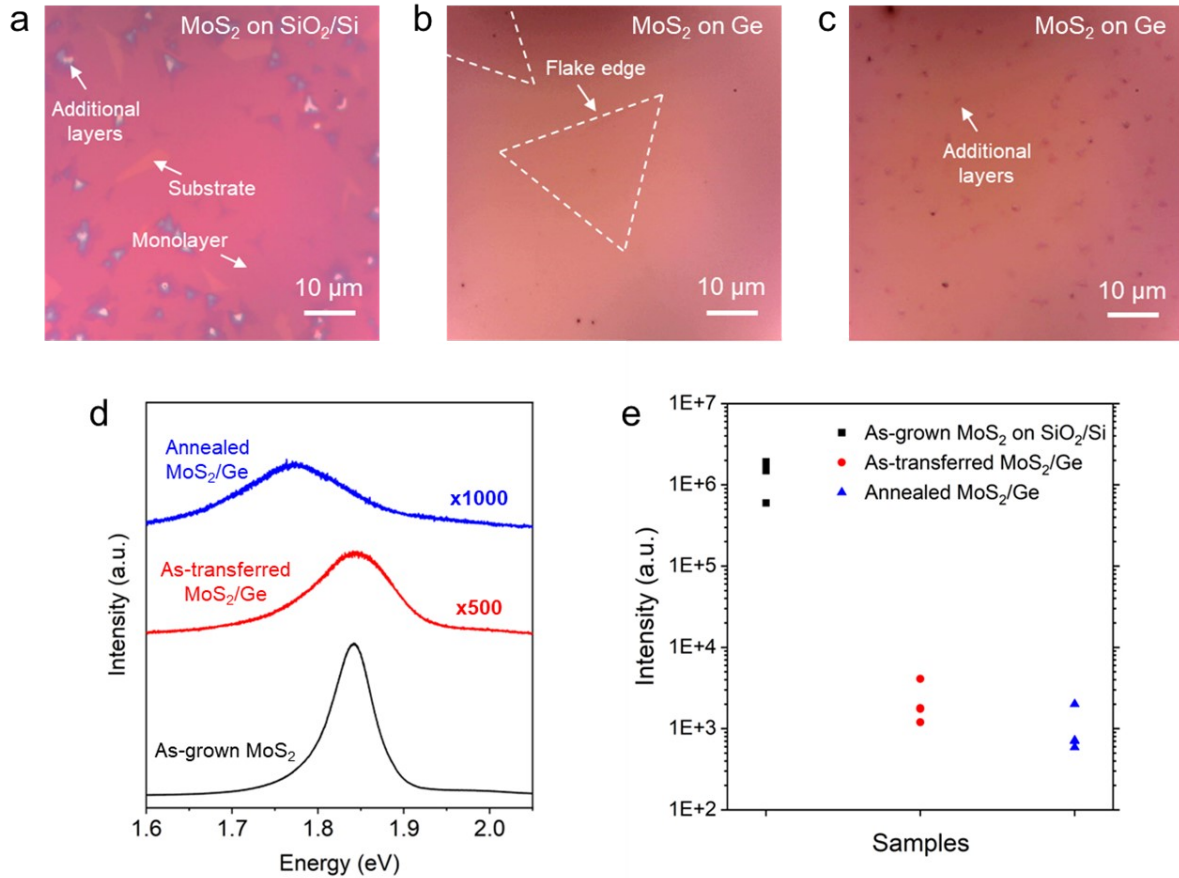


Figure S3. PL studies of MoS₂/Ge heterostructures and effects of post-transfer annealing, in which the monolayer MoS₂ is grown on SiO₂/Si by CVD and then transferred to Ge. (a) Optical image of as-grown monolayer MoS₂ on SiO₂/Si. The monolayer MoS₂, bare substrate, and additional MoS₂ layer regions are distinguished by optical contrast and marked by white arrows. (b) Optical image of typical MoS₂ flakes transferred on Ge. (c) Optical image of continuous films of monolayer MoS₂ on Ge. MoS₂ monolayers exhibit low optical contrast on Ge. (d) A comparison of PL spectra of as-grown monolayer MoS₂, as-transferred MoS₂/Ge, and annealed MoS₂/Ge. MoS₂/Ge heterostructures display prominent PL intensity quenching compared to as-grown MoS₂. (e) A plot of PL intensity distributions of the samples investigated in (d). For each case, four random spots are measured, so that the spatial variation of PL intensity is taken into account.

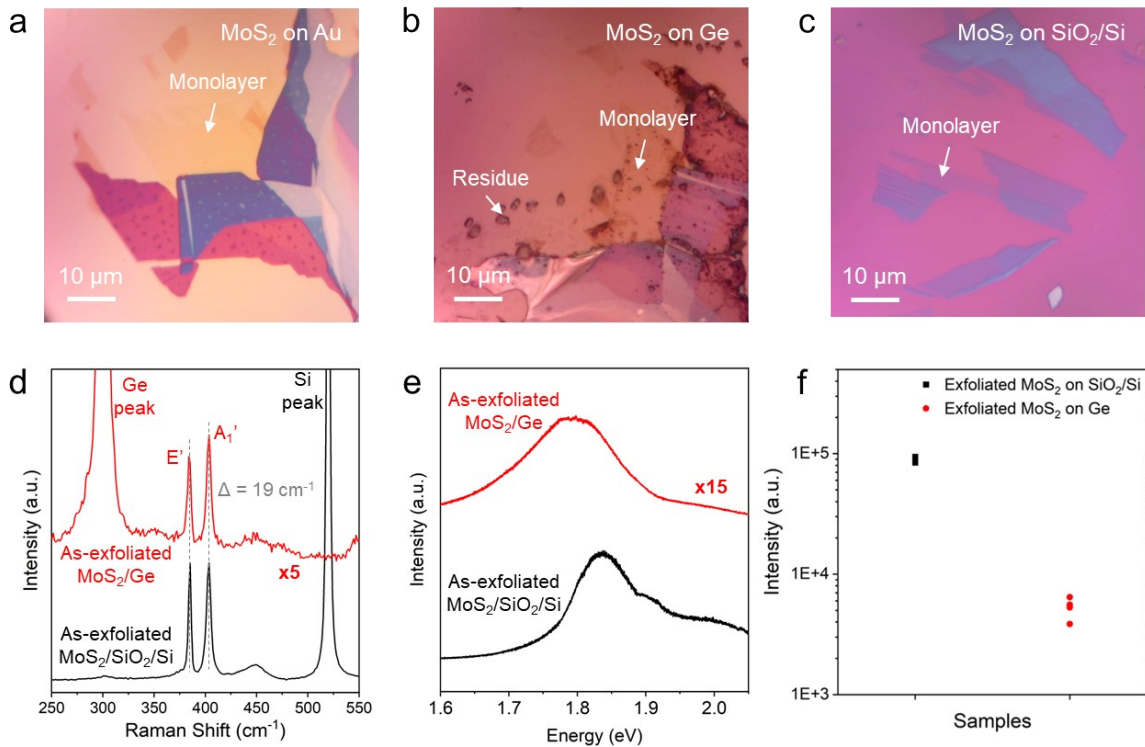


Figure S4. PL studies of MoS₂/Ge heterostructures obtained by exfoliating monolayer MoS₂ on Ge. Considering the very low optical contrast of TMD monolayers on Ge, a previously reported gold (Au)-assisted approach was adopted to increase the exfoliation yield and aid the identification of monolayers.¹ **(a)** Optical image of a MoS₂ flake exfoliated on a gold tape (consisting of Au(150 nm)/PMMA/thermal release tape (TRT)), in which the monolayer region can be distinguished based on optical contrast. **(b)** Optical image of the identical flake transferred onto Ge. The monolayer MoS₂ region can be determined by comparing this optical image with (a). **(c)** Optical image of a monolayer MoS₂ exfoliated on SiO₂/Si. **(d)** Raman spectra of exfoliated MoS₂/SiO₂/Si and MoS₂/Ge. The monolayer nature of these flakes can be verified by the separation ($\sim 19\text{ cm}^{-1}$) between E' and A₁' Raman modes.^{2,3} **(e)** A comparison of PL spectra of exfoliated MoS₂/SiO₂/Si and MoS₂/Ge. It should be noted that we were not able to obtain the PL spectra of annealed MoS₂/Ge, as our current exfoliation approach resulted in surface residue on both the MoS₂ flakes and Ge substrate. The residue seemed to be aggregated on top of the MoS₂ flake during the annealing process, hindering further optical characterizations. Future work is required to improve

our exfoliation technique in order to study the exfoliated MoS₂/Ge heterostructures in detail. **(f)** A plot of PL intensity distributions of the samples investigated in (e). For each case, four random spots are measured, so that the spatial variation of PL intensity is taken into account.

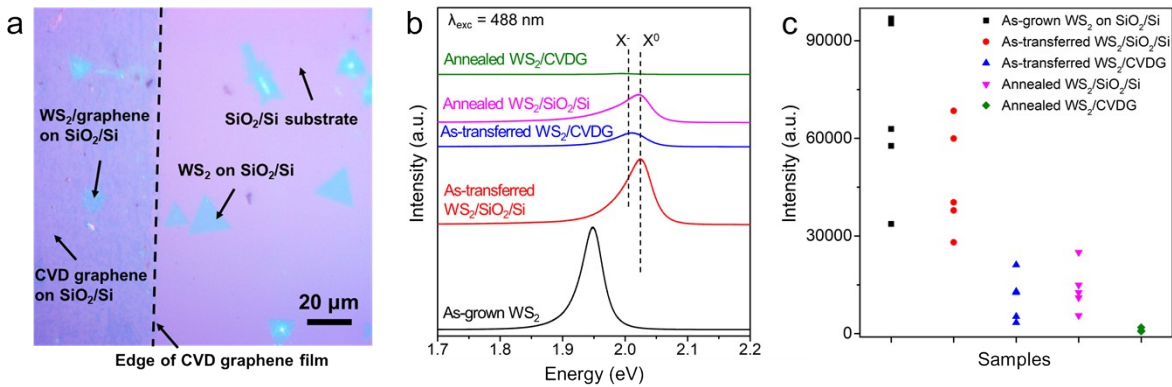


Figure S5. Effects of post-transfer annealing on the PL intensity and uniformity of WS₂/SiO₂/Si and WS₂/CVD-grown monolayer graphene (CVDG)/SiO₂/Si heterostructures.

(a) Optical image of monolayer WS₂ transferred on CVDG/SiO₂/Si. The left side of the SiO₂/Si substrate is covered with CVDG (purple colored region) while the right side of the substrate is without CVDG. Thus, both WS₂/SiO₂/Si and WS₂/CVDG/SiO₂/Si can be found and measured within this sample. **(b)** A comparison of PL spectra of as-grown monolayer WS₂, as-transferred WS₂/SiO₂/Si, annealed WS₂/SiO₂/Si, as-transferred WS₂/CVDG/SiO₂/Si, and annealed WS₂/CVDG/SiO₂/Si. **(c)** A plot of PL intensity distributions of the samples investigated in (b). For each case, five random spots are measured, so that the spatial variation of PL intensity is taken into account. The results indicate a consistent PL intensity quenching and uniformity improvement after the low-pressure Ar/H₂ annealing process.

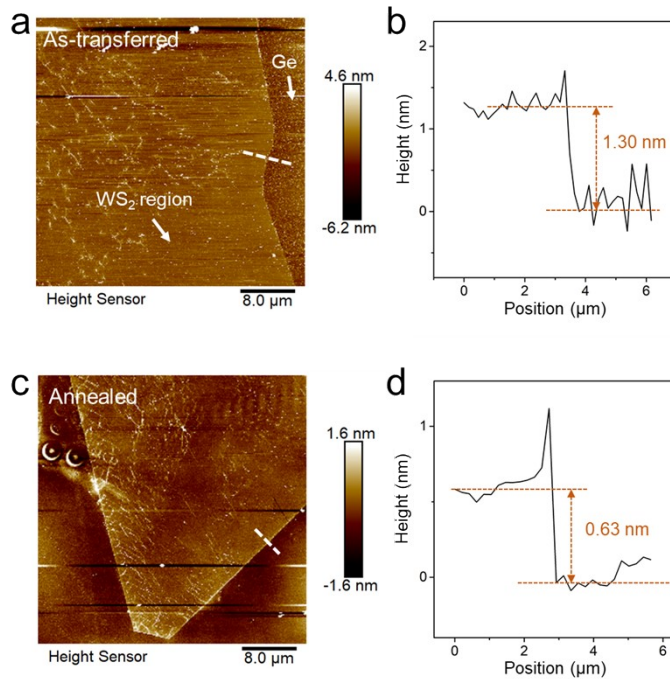
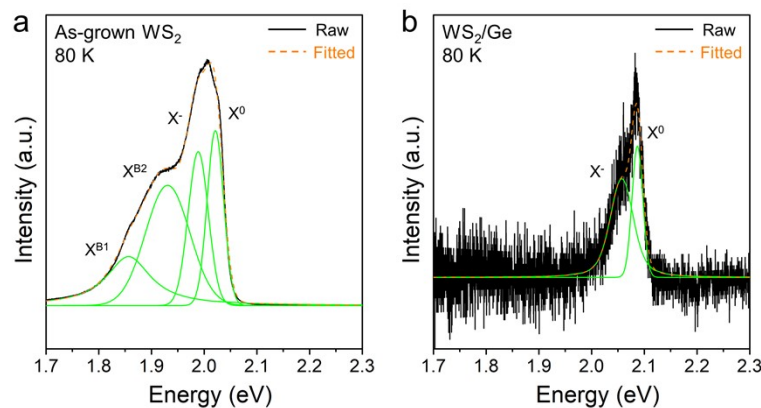


Figure S6. Additional AFM data of as-transferred and annealed WS₂/Ge heterostructures, which were obtained from different flakes with the data presented in Figures 2e-h. (a) AFM topography image of as-transferred WS₂/Ge and **(b)** the corresponding surface height profile along the white dashed line in (a). **(c)** AFM topography image of annealed WS₂/Ge and **(d)** the corresponding surface height profile along the white dashed line in (c). The reduction of WS₂/Ge



distance is consistently observed.

Figure S7. PL spectra of as-grown WS₂ and WS₂/Ge heterostructures measured at 80 K. The spectra were deconvoluted and exciton species were determined by peak fitting using Voigt

profiles. Individual peaks are plotted with green solid lines, while the cumulative fitted spectra are plotted with orange dashed lines.

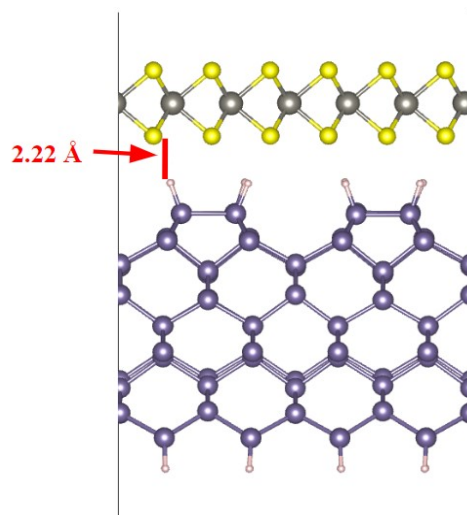


Figure S8. DFT-calculated structure of WS₂/H-Ge(100). The distance between the topmost H atoms and topmost S atoms is calculated to be 2.22 Å.

References

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