Supplementary information

Characterization of the structural defects of CVD-grown monolayer MoS₂

using near-field photoluminescence imaging

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Figure S1. Average PL and Raman spectra of the monolayer MoS_2 flakes. (a) Confocal PL spectrum and (b) Raman spectrum obtained from a monolayer MoS_2 flake. The fitted Raman peak separation between the E_{2g}^1 and A_{1g} Raman peaks was measured to be 17.4 cm⁻¹. This value is in the range 17.0~19 cm⁻¹ which is the range of reported values of Raman peak separation of monolayer MoS_2^{1-5} . We also checked the Raman peak separation of monolayer MoS_2 before and after the transfer process because strain could be introduced during the transfer process⁶. However, we didn't find any notable change before and after the transfer process.



Figure S2. Correlated SEM and SNOM PL images of a monolayer MoS_2 flake.(a) SEM and (b) SNOM PL images of a monolayer MoS_2 flake. Note that the line defects in the SEM image were observed in the SNOM PL image. The scale bar indicates 3 μ m.



Figure S3. Spatial resolution of the SNOM PL and confocal PL images. (a) Confocal PL image and (b) SNOM PL image of monolayer MoS₂. (c) Line profiles of a single region obtained from the confocal PL images and SNOM PL images indicated in (a, b) by the red and blue lines. The spatial resolutions were measured based on the distance between the positions corresponding to 12% and 88% of the PL intensity. The spatial resolutions of the SNOM PL and confocal PL images were estimated to be 110 nm and 440 nm, respectively. (d) AFM image of an Al pattern. (e) Optical transmission image of the Al pattern obtained using the same SNOM instrument. (The AFM and SNOM images were not from the same sample area). The scale bars in (a, b) indicate 6 μ m; the scale bars in (d, e) indicate 1 μ m, and the scale bars in the insets indicate 200 nm.



Figure S4. SNOM PL, confocal PL, and Raman mapping images of monolayer MoS₂ flakes prepared on the sapphire substrate.

(a, e) SEM image, (b, f) SNOM PL images, (c, g) confocal PL intensity images, and
(d, h) Raman intensity mapping images of monolayer MoS₂ flakes. The scale bars in
(a) indicate 2 μm, and (e) 1 μm.



Figure S5. SNOM PL images of polycrystalline monolayer MoS_2 flakes.

(**a**, **b**) SNOM PL images showing the triangular-shaped adlayer defects. Thirty adlayer defects were selected from the center regions of two MoS_2 flakes (red boxed regions), and their relative orientations to the underlying MoS_2 grain were measured (See the main text). The scale bars in the main images indicate 5 µm and the scale bars in the insets indicate 1 µm.



Figure S6. SNOM PL and confocal PL images of polycrystalline monolayer MoS₂ flakes.

(**a**, **b**) SNOM and confocal PL images of monolayer MoS₂ which contains a mirrortype GB. No PL reduction on the center line, where GB is expected to exist, is observed (**c**, **d**) Confocal PL images of monolayer MoS₂ which has multiple GBs. No clear PL contrast along GB-expected locations is observed. Possible GB positions are indicated with white arrows.



Figure S7. SNOM and confocal PL images of defects in monolayer MoS₂. (a) SNOM PL image of monolayer MoS₂, kept in the air for days. Fresh-cut region as marked yellow dotted circle didn't show PL enhancement, while some indication of PL increase along the edge is noticed, probably due to environmental oxidation and resulting p-type doping. (b) Confocal PL image of monolayer MoS₂, kept in the air for days. PL enhancement was detected along the edge and a line defect edge. The scale bars indicate 3 μm.



Figure S8. SNOM PL image pairs. (a, b), (c, d) SNOM PL image pairs of monolayer MoS_2 in same region. a and c image were obtained first, and then b and d were obtained again. No degradation of PL images by the laser irradiation is observed.

Reference

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