

ADVANCED FUNCTIONAL MATERIALS

Supporting Information

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Electrochemically Exfoliating MoS₂ into Atomically Thin
Planar-Stacking Through a Selective Lateral Reaction
Pathway

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Supplementary Methods

Fitting of Raman spectra. The Raman spectra of MoS₂ and MoSe₂ are fitted by LabSpec 6 software. Firstly remove baseline polynomial fitting and then search peaks and fit by Gauss-Lorentz line shape. The Gauss-Lorentz line shape is described as: $V(\nu-\nu_0)=\int G(\nu'_0-\nu_0)L(\nu-\nu'_0)d\nu'_0$, $G(\nu-\nu_0)=A\exp\frac{-(\nu-\nu_0)}{2(\omega/2)^2}$, $L(\nu-\nu_0)=\frac{\omega/2\pi}{(\nu-\nu_0)^2+(\omega/2)^2}$. where ν is Raman shift, ω is the full width at half maximum (FWHM), ν_0 is the position of the peak. The wavenumber difference between E_{2g}¹ (~383 cm⁻¹) and A_{1g} (~408 cm⁻¹) of MoS₂ is calculated by $\Delta\nu = \nu_{A_{1g}} - \nu_{E_{2g}^1}$.

Supplementary Figures

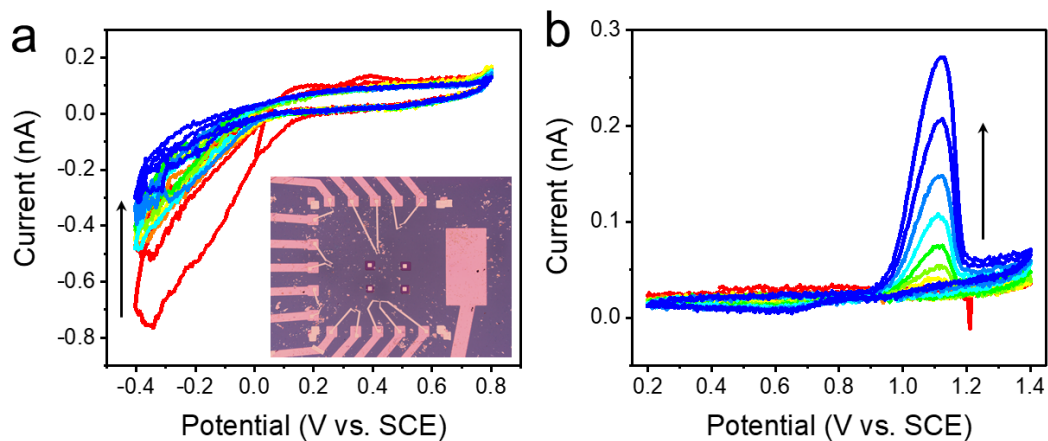


Figure S1. (a) Cyclic voltammetry (CV) curves of individual MoS₂ sheet in 1 M ZnSO₄ solution (first 10 cycles, -0.4 - 0.8 V vs. SCE, scan rate 10 mV/s). Inset: the optical image of a typical on-chip device. (b) CV curves of individual MoS₂ sheet in 1 M ZnSO₄ solution at the oxidation region (first 10 cycles, 0.2 - 1.4 V vs. SCE, scan rate 50 mV/s).

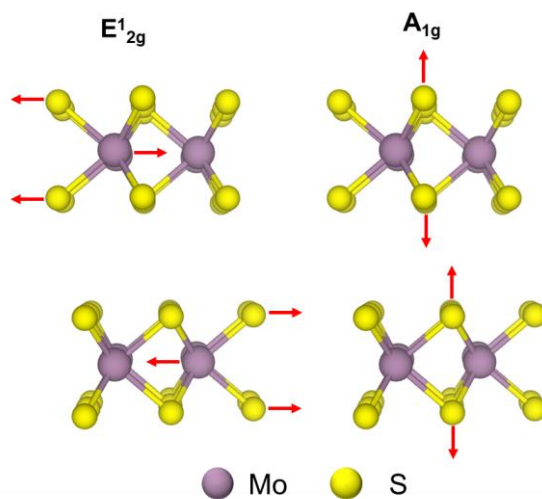


Figure S2. Two characteristic Raman mode for 2H-MoS₂. E_{12g}¹ is the in-plane opposite Mo-S vibration and A_{1g} is the out-of-plane vibration of S atoms.

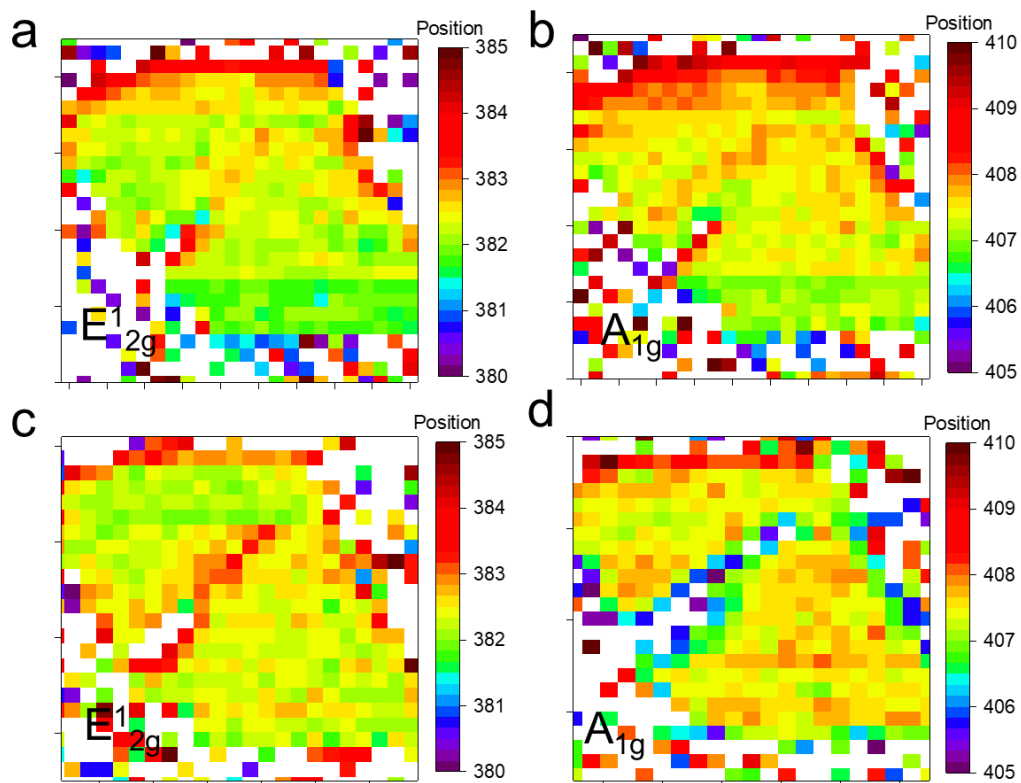


Figure S3. (a,b) Raman mapping of E_{2g}^1 and A_{1g} vibration bands for the initial MoS_2 sheet corresponding to the MoS_2 in Figure 2a. (c,d) Raman mapping of E_{2g}^1 and A_{1g} vibration bands for MoS_2 sheet after electrochemical treatment corresponding to the MoS_2 sheet in Figure 2c. The color represents the wavenumber of E_{2g}^1 and A_{1g} peaks.

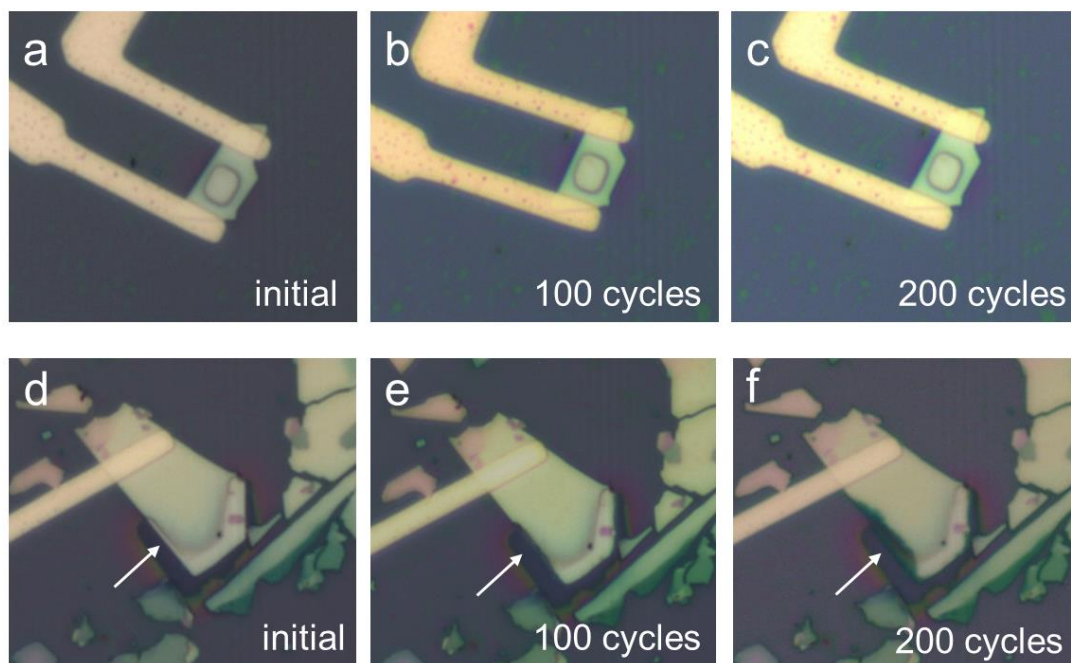


Figure S4. (a-c) The optical images of individual MoS₂ with basal plane exposed and edge cover by PMMA. (c) and (d) are optical images of MoS₂ after 100 and 200 CV cycles. (d-f) The corresponding optical images of individual MoS₂ with edge exposed. The arrow points to the reaction region.

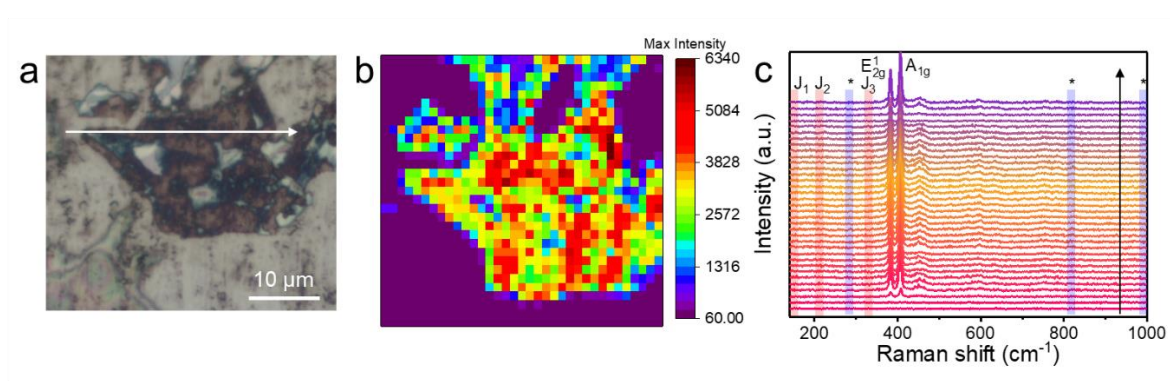


Figure S5 (a) Optical image of a MoS₂ sheet after electrochemical exfoliation. (b) Raman mapping of the intensity of A_{1g} mode. (c) The selected Raman spectra along the line in (a). The peak positions of J₁, J₂, and J₃ are marked by red bands. The peak positions of MoO_x are marked by asterisks and blue bands.

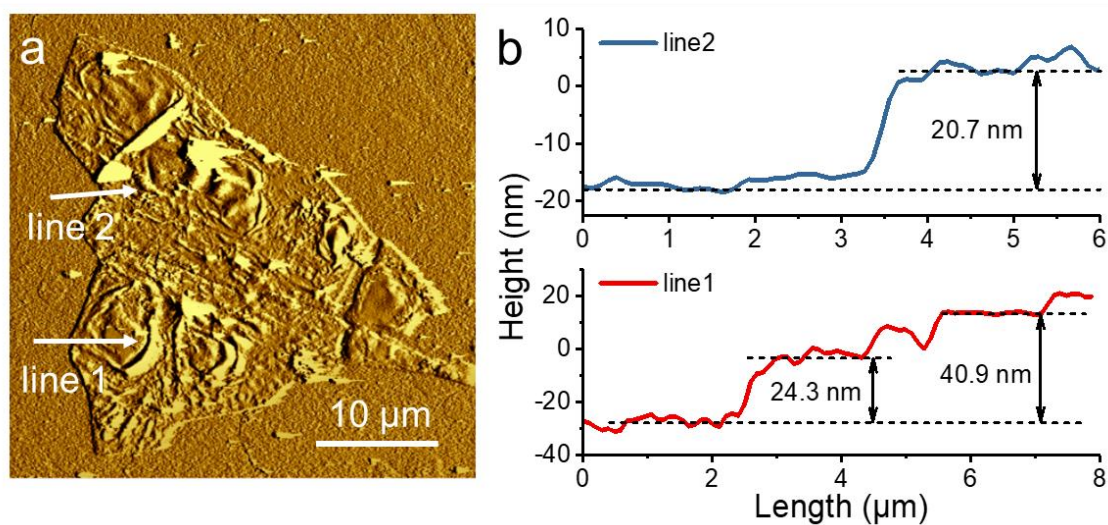


Figure S6. (a) AFM image of a typical MoS₂ nanosheet corresponding to the MoS₂ sheet in Figure 4b after electrochemical treatment on Ti foil with Si wafer as the substrate. (b) The height profile along line 1 and line 2.

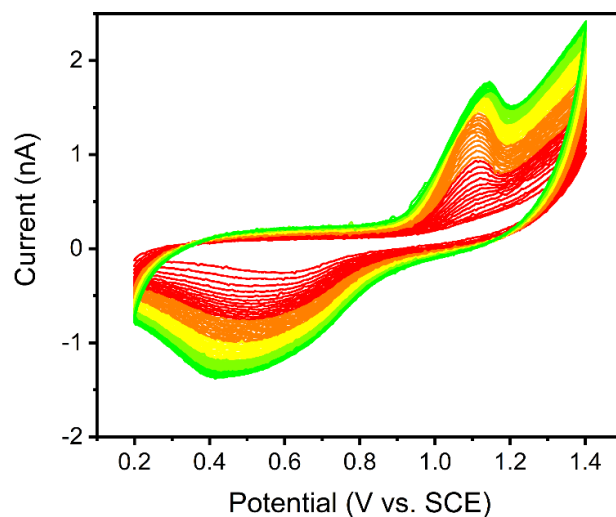


Figure S7. First tens of CV curves of individual MoSe₂ sheet in 1M ZnSO₄ (scan window: 0.2-1.4 V vs. SCE, scan rate: 50 mV/s).

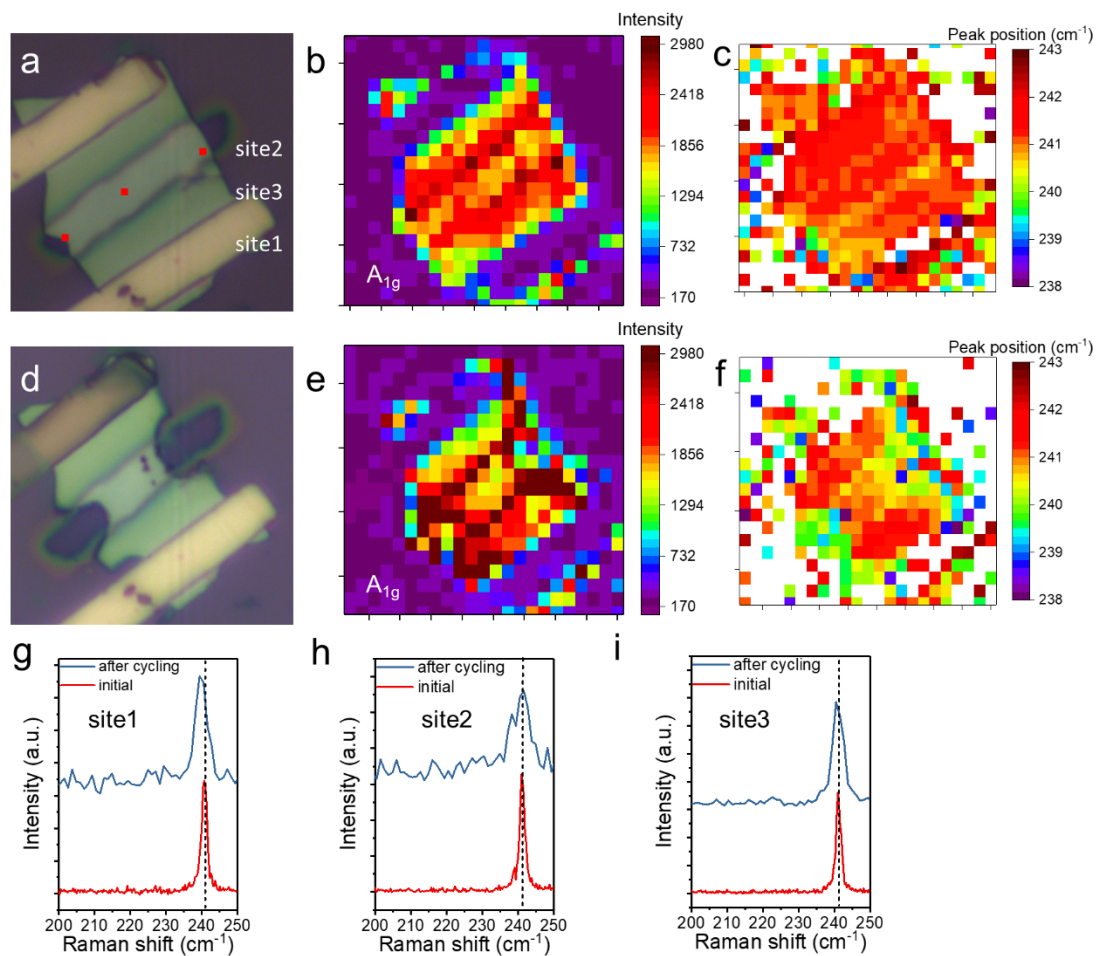


Figure S8. (a) Optical image of individual MoSe₂ sheet on-chip device with PMMA as the insulating layer. (b) Raman mapping of the intensity and (c) wavenumber for the A_{1g} vibration band. (d) The optical image, (e) Raman mapping of intensity, and (f) wavenumber for MoSe₂ sheet after electrochemical treatment. (g-i) Raman spectra of three selected points in (a).

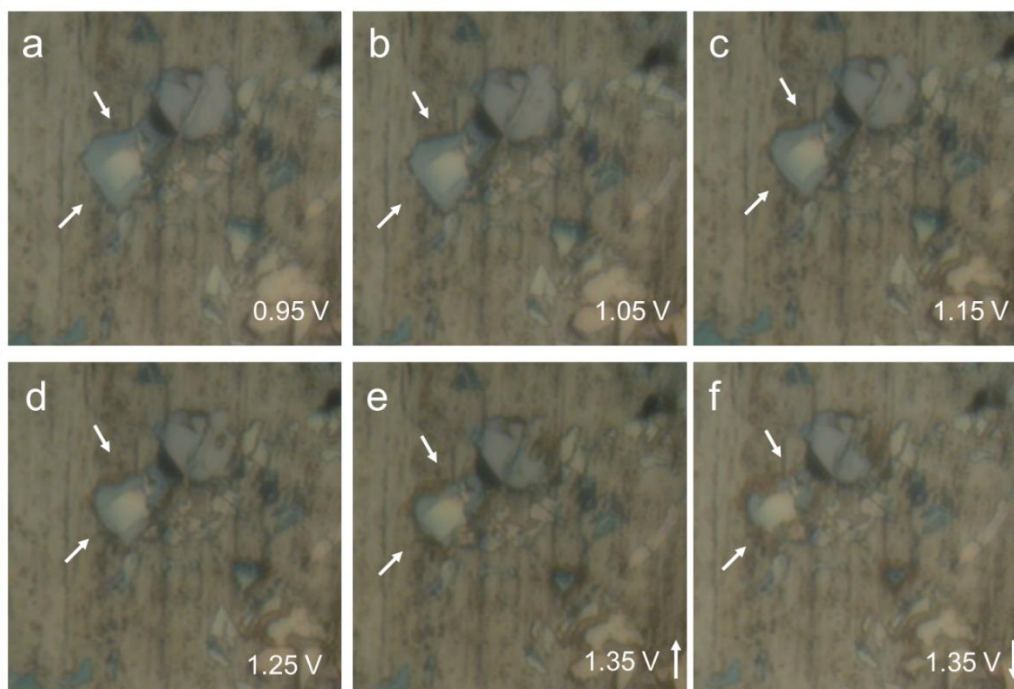


Figure S9. (a-f) The *in situ* optical snapshots of MoSe₂ sheets on Ti foil during an electrochemical process in the ZnSO₄ electrolyte (electrode potential first increases to 1.4 V vs. SCE and then back). It is worth noting that (e) is the image of MoSe₂ at 1.35 V during the anodic process and (f) is at 1.35 V during the cathodic process.

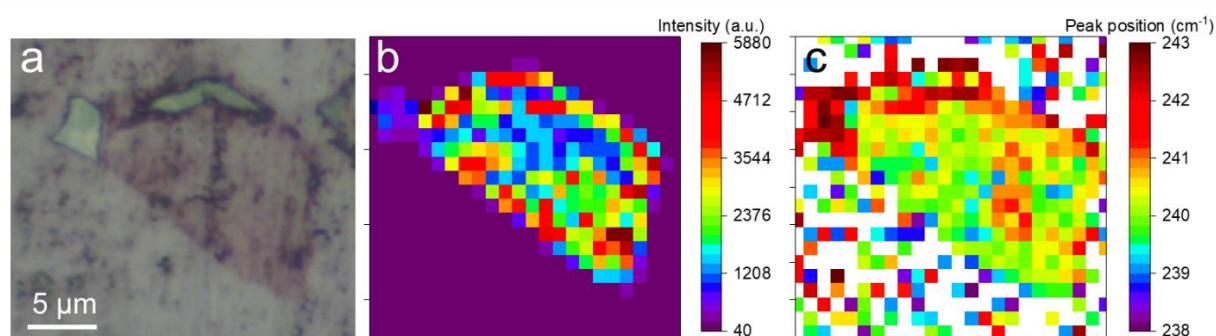


Figure S10. (a) Optical image of MoSe₂ sheet on Ti foil after electrochemical treatment. The transparent trace shows the left ultrathin MoSe₂ on Ti foil. (c) The Raman mapping of the intensity of the A_{1g} vibration band. (d) The corresponding mapping of the wavenumber of the A_{1g} vibration band.

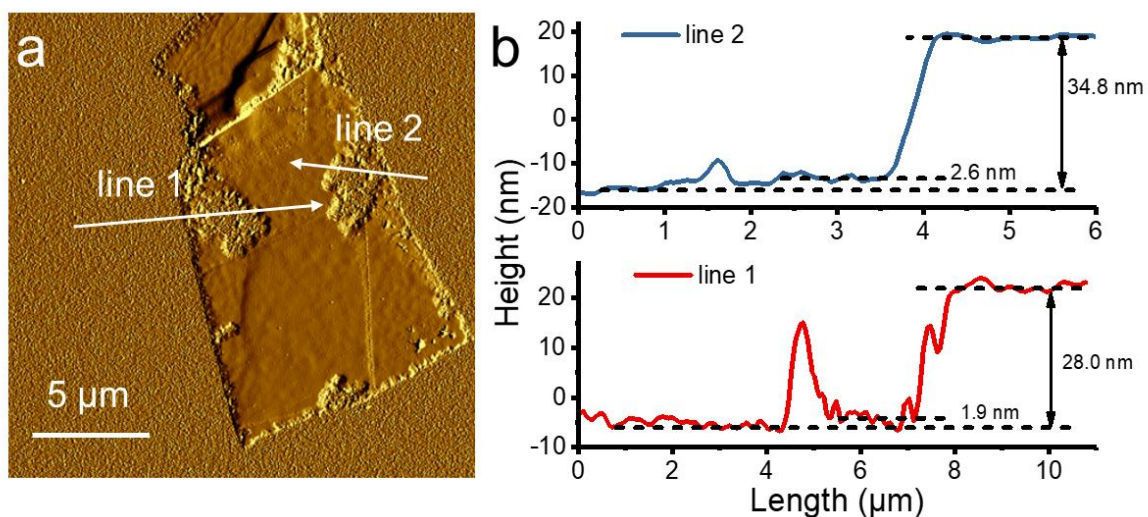


Figure S11. (a) AFM image of a typical MoSe₂ nanosheet after electrochemical treatment on Ti foil with Si wafer as a substrate. (b) The corresponding height profile along line 1 and line 2.

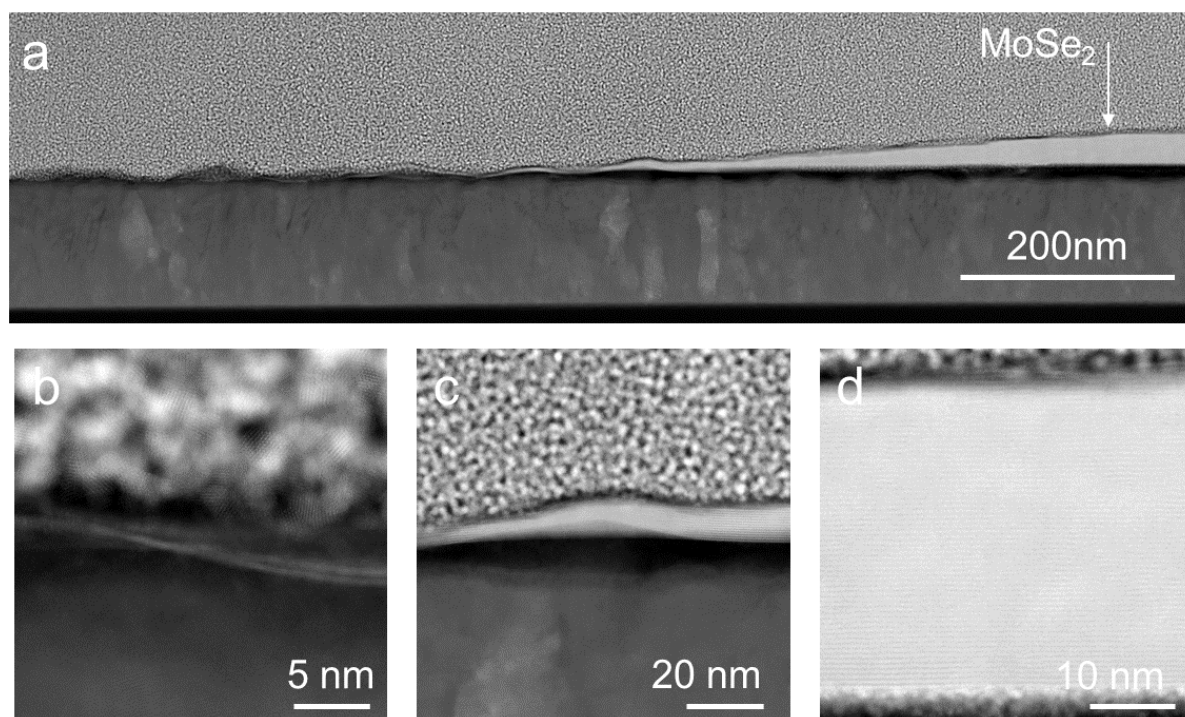


Figure S12. (a) An overview of the cross-profile STEM image of the exfoliated MoSe₂ sheet (b-d) The magnified STEM images of edge, transition zone, and bulk, respectively.

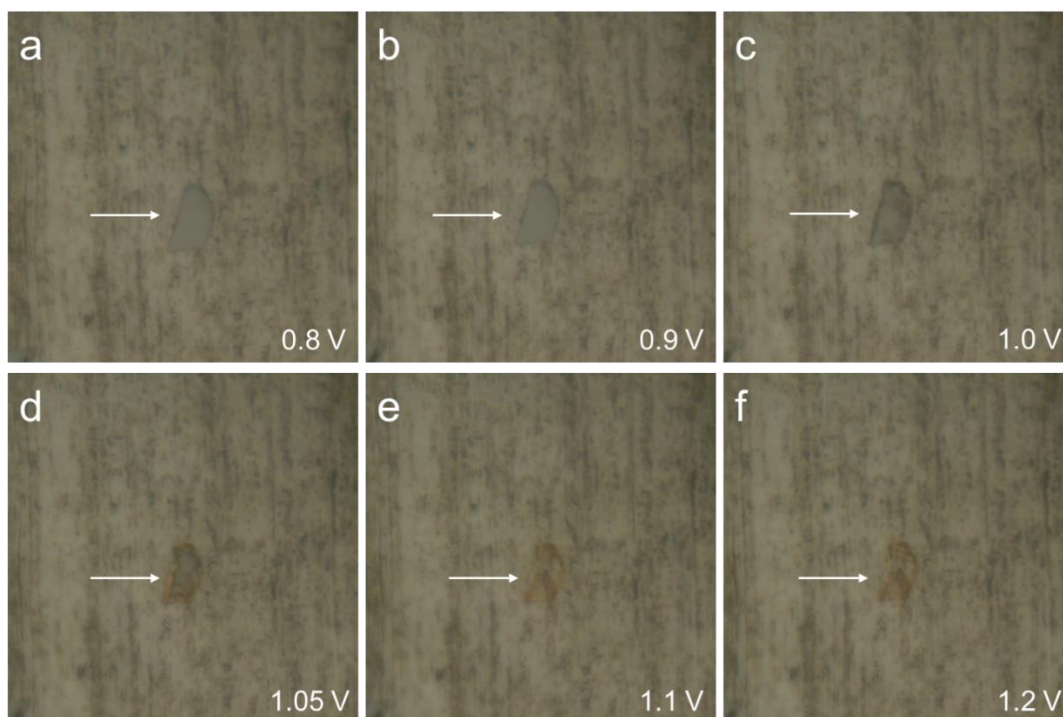


Figure S13. (a-f) Optical images of VSe_2 on Ti foil during an anodic process at 0.8, 0.9, 1.0, 1.05, 1.1, and 1.2 V vs. SCE in 1M ZnSO_4 . The white arrows point to an individual VSe_2 sheet on Ti foil.

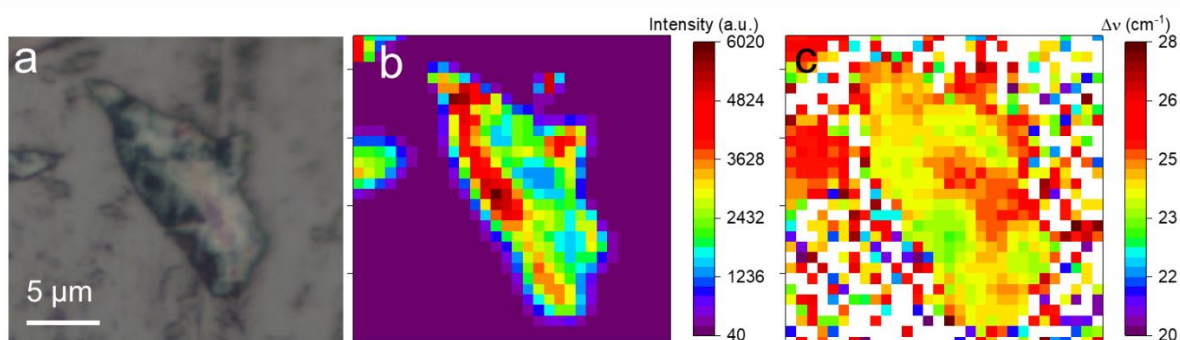


Figure S14. (a) Optical image of MoS_2 sheet on Ti foil after electrochemical treatment in the 1M Na_2SO_4 solution. The left of this sheet in green color is the domain involved in exfoliation. (c) The Raman mapping of the sum intensity of E_{2g}^1 and A_{1g} vibration bands. (d) The corresponding mapping of the wavenumber difference between E_{2g}^1 and A_{1g} vibration

bands.

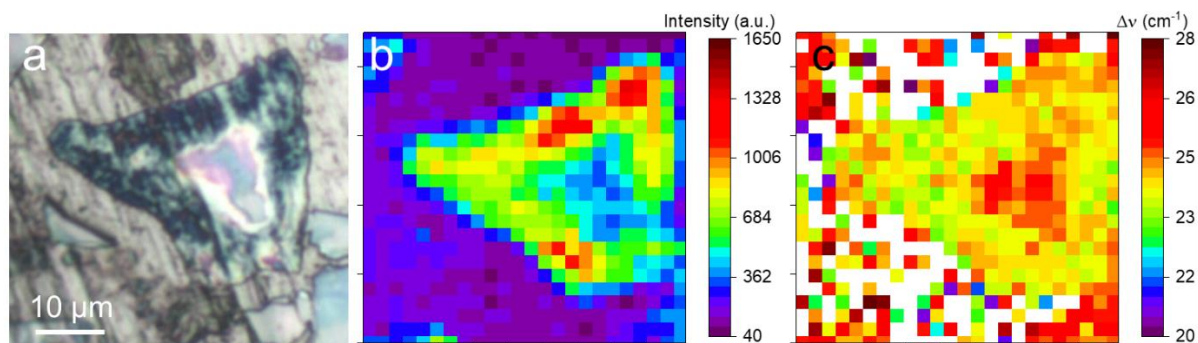


Figure S15. (a) Optical image of MoS₂ sheet on Ti foil after electrochemical treatment in the 1M Li₂SO₄ solution. (c) The Raman mapping of the sum intensity of E_{12g}¹ and A_{1g} vibration bands. (d) The corresponding mapping of the wavenumber difference between E_{12g}¹ and A_{1g} vibration bands.

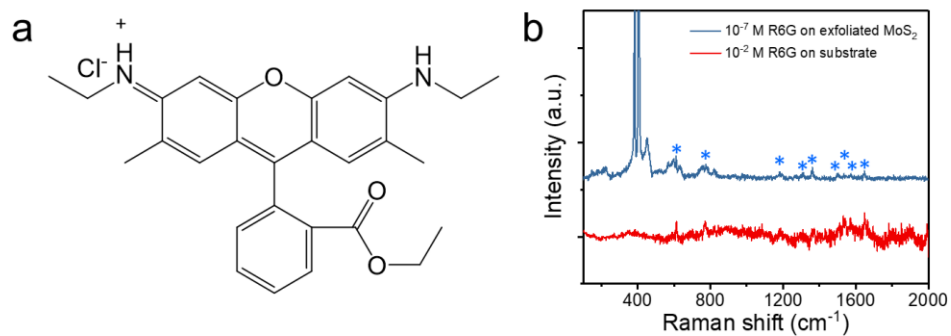


Figure S16. (a) Structure of R6G molecule. (b) Raman spectra of R6G on exfoliated MoS₂ and Ti foil substrate. The enhancement factor is calculated through the intensity of peaks at 613 cm⁻¹.