Supporting Information


Electrochemically Exfoliating MoS$_2$ 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 MoS\textsubscript{2} and MoSe\textsubscript{2} 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:

\[ V(\nu - \nu_0) = \int G(\nu' - \nu_0) L(\nu' - \nu_0) d\nu_0 , \]

\[ G(\nu' - \nu_0) = A \exp\left(\frac{- (\nu' - \nu_0)^2}{2 \omega^2}\right) , \]

\[ L(\nu_0) = \frac{\omega}{\pi} \left(\frac{\nu' - \nu_0}{\nu_0^2 + (\omega/2)^2}\right) . \]

where \( \nu \) is Raman shift, \( \omega \) is the full width at half maximum (FWHM), \( \nu_0 \) is the position of the peak. The wavenumber difference between \( E_{2g}^{1} \approx 383 \text{ cm}^{-1} \) and \( A_{1g} \approx 408 \text{ cm}^{-1} \) of MoS\textsubscript{2} is calculated by \( \Delta \nu = \nu_{A_{1g}} - \nu_{E_{2g}^{1}} \).
Supplementary Figures

**Figure S1.** (a) Cyclic voltammetry (CV) curves of individual MoS$_2$ sheet in 1 M ZnSO$_4$ 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 MoS$_2$ sheet in 1 M ZnSO$_4$ 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-MoS$_2$. $E_{2g}^1$ 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 MoS$_2$ with basal plane exposed and edge cover by PMMA. (c) and (d) are optical images of MoS$_2$ after 100 and 200 CV cycles. (d-f) The corresponding optical images of individual MoS$_2$ with edge exposed. The arrow points to the reaction region.

Figure S5 (a) Optical image of a MoS$_2$ sheet after electrochemical exfoliation. (b) Raman mapping of the intensity of A1g mode. (c) The selected Raman spectra along the line in (a). The peak positions of $J_1$, $J_2$, and $J_3$ 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 MoS$_2$ nanosheet corresponding to the MoS$_2$ 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 MoSe$_2$ sheet in 1M ZnSO$_4$ (scan window: 0.2-1.4 V vs. SCE, scan rate: 50 mV/s).
Figure S8. (a) Optical image of individual MoSe$_2$ 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 MoSe$_2$ sheet after electrochemical treatment. (g-i) Raman spectra of three selected points in (a).
Figure S9. (a-f) The *in situ* optical snapshots of MoSe$_2$ sheets on Ti foil during an electrochemical process in the ZnSO$_4$ electrolyte (electrode potential first increases to 1.4 V vs. SCE and then back). It is worth noting that (e) is the image of MoSe$_2$ 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 MoSe$_2$ sheet on Ti foil after electrochemical treatment. The transparent trace shows the left ultrathin MoSe$_2$ 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 MoSe$_2$ 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 MoSe$_2$ 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$_2$SO$_4$ 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 MoS$_2$ sheet on Ti foil after electrochemical treatment in the 1M Li$_2$SO$_4$ solution. (c) The Raman mapping of the sum intensity of E$_{1\,2g}$ and A$_{1g}$ vibration bands. (d) The corresponding mapping of the wavenumber difference between E$_{1\,2g}$ and A$_{1g}$ vibration bands.

**Figure S16.** (a) Structure of R6G molecule. (b) Raman spectra of R6G on exfoliated MoS$_2$ and Ti foil substrate. The enhancement factor is calculated through the intensity of peaks at 613 cm$^{-1}$. 