Supplementary Information

Direct Electron-beam Patterning of Monolayer MoS₂ with Ice

Guangnan Yao^{1,2,3}, Ding Zhao^{2,3,*}, Yu Hong¹, Shan Wu^{1,2,3}, Dongli Liu^{2,3}, and Min Qiu^{2,3,*}

 College of Optical Science and Engineering, Zhejiang University, Hangzhou 310027, China

2. Key Laboratory of 3D Micro/Nano Fabrication and Characterization of Zhejiang Province, School of Engineering, Westlake University, Hangzhou 310024, China

3. Institute of Advanced Technology, Westlake Institute for Advanced Study, Hangzhou 310024, China

*E-mail: zhaoding@westlake.edu.cn (D.Z.), qiu_lab@westlake.edu.cn (M.Q.)

Influence of ice on MoS₂



Figure S1. Raman mapping of MoS_2 (a-b) before and (c-d) after ice deposition followed by sublimation. E-beam exposure was not performed.

Honeycomb structures



Figure S2. SEM image of honeycomb structures on sample surface after e-beam exposure. The e-beam dose is 4 C/cm^2 at 10 keV.

Raman spectral mapping



Figure S3. Raman mapping of the patterned MoS_2 flake in Figure 3 at characteristic peaks of (a) 384.1 cm⁻¹ and (b) 403.1 cm⁻¹, respectively.

Patterning of monolayer MoSe₂



Figure S4. (a) Raman mapping of monolayer $MoSe_2$ on a SiO_2 (300 nm)/Si substrate at the characteristic peak, where $1.5\mu m \times 1.5\mu m$ squares were patterned. The dose of each square is same as that in Figure 3; (b) Raman spectra obtained within unexposed (red line) and exposed areas (blue line) in (a).

Patterning on a free-standing sample

We tried patterning a free-standing monolayer MoS_2 on TEM copper nets instead of SiO_2/Si substrates. Compared with bulk substrates, less backscattered electrons were produced in free-standing samples, thus the MoS_2 cannot be removed effectively. An energy dispersive spectrometer was used to characterize elemental composition of the patterned area, and both Mo and S existed on the sample (data not shown).



Figure S5. TEM image of free-standing MoS_2 after ice-assisted patterning in our instrument. The area within the red square was irradiated by a 10 keV e-beam with dose of 4 C/cm².