Supporting Information

Controllable Fabrication and Photocatalytic Performance of Nanoscale Single-layer $MoSe_2$ with Substantial Edges on Ag(111)

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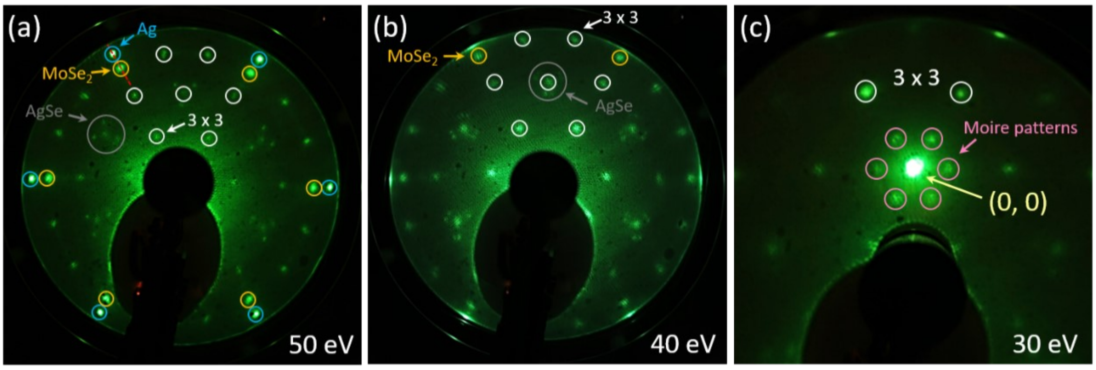

Figure S1. (a) LEED patterns of SL-$MoSe_2$ islands on the Ag(111) substrate at 50 eV (a), 40 eV (b) and 30 eV (c). Diffraction spots marked by blue cycles, yellow cycles and pink cycles are from Ag(111) substrate, $MoSe_2$ and moiré pattern, respectively. The diffraction spots marked by gray cycles are belong to the AgSe monolayer on the Ag(111) substrate. 3 × 3 diffraction spots with respect to Ag(111) substrate (marked by white cycles) may come from unknown silver selenide compound.

Besides the diffraction spots from Ag(111) and $MoSe_2$ in Figure S1a, the rest spots are attributed to the moiré patterns and silver selenide compounds. Figure S1c shows a LEED pattern by rotating the sample for an angle to expose the zero-order diffraction spot. The surrounded six spots of zero order (highlighted by six pink cycles) come from the moiré superlattice, which is in good agreement with the experimental observations. The diffraction spots marked by gray cycles in Figure S1a and 1b are the typical LEED patterns of the AgSe monolayer on Ag(111) substrate[1]. In addition, for the 3 × 3 diffraction spots (marked by white cycles) with respect to Ag substrate, we predict it would be some kind of silver selenide compounds.

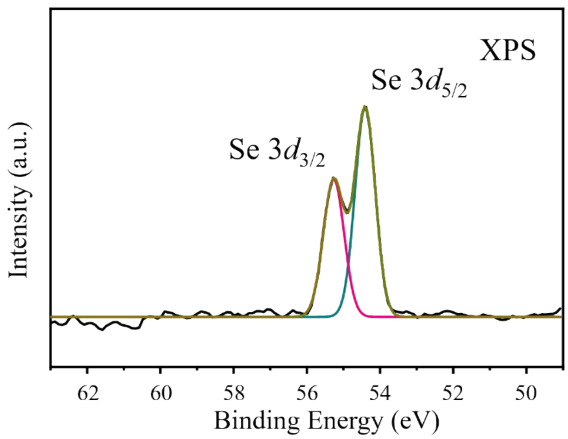

Figure S2. Se XPS spectrum of SL-$MoSe_2$ islands on the Ag(111) substrate after subtracting the Ag 4p core level.

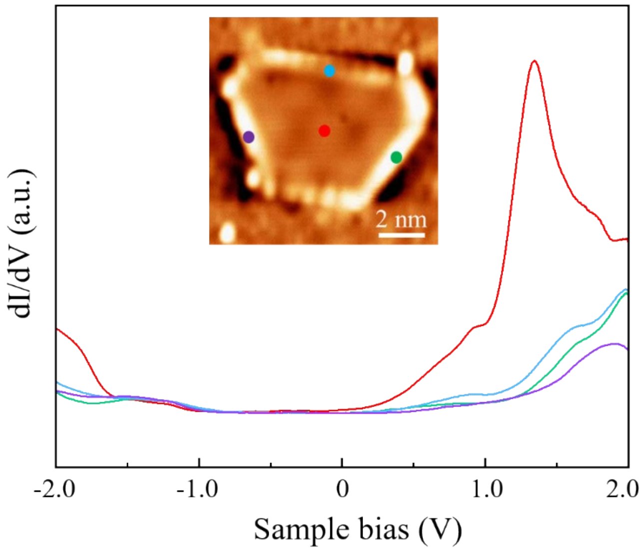

Figure S3. Four dI/dV curves taken on the three edges of one SL-$MoSe_2$ island and inside the island on the Ag(111) substrate (inset STM image).

We also performed STS measurements on the edge of SL-$MoSe_2$ islands on the Ag(111) substrate. Figure S3 presents four dI/dV curves taken on a $MoSe_2$ island. Three of them collected from the three longer edges of the $MoSe_2$ island (marked by blue, green, and purple) have the same variation trend with the curves measured on the longer edges of SL-$MoSe_2$ islands on Au(111) substrate.[2] Therefore, we predict that the as-synthesized SL-$MoSe_2$ islands on the Ag(111) substrate are preferentially terminated by Mo edges with single Se atoms saturated in longer ones and bare Se edges in shorter ones.

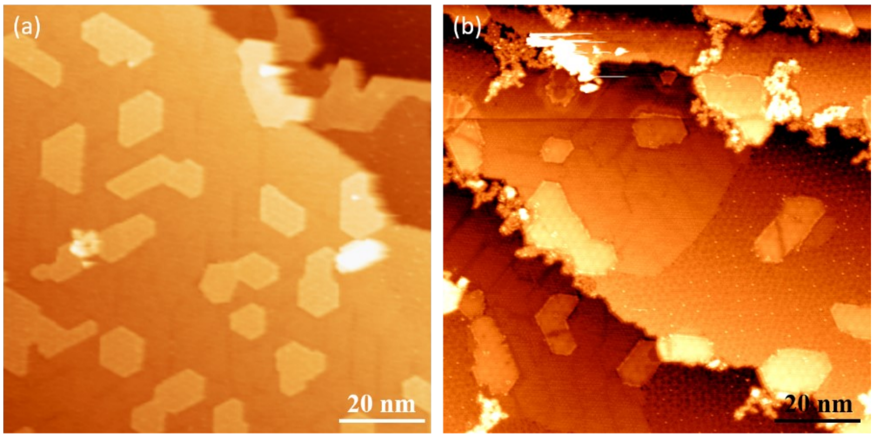

Figure S4. (a) STM image of SL-$MoSe_2$ islands on the Ag(111) substrate after annealing 450 °C for 30 min. $V_s = -3.0$ V, $I_t = 0.03$ nA. (b) STM image of SL-$MoSe_2$ islands on the Ag(111) substrate to prove the stability in ambient conditions. $V_s = -1.0$ V, $I_t = 0.1$ nA.

The nanoscale SL-$MoSe_2$ islands on Ag(111) substrate has excellent thermal stability. As shown in Figure S4a, after annealing to 450 °C for 30 min, the nanoscale single-layer $MoSe_2$ islands still maintain their original structures. In addition, the nanoscale SL-$MoSe_2$ islands are stable in ambient conditions at room temperature. To demonstrate this property, the sample was removed from the vacuum chamber and was kept in air without protection for more than 2 h. Then the sample was put back to the vacuum chamber and annealed at 450 °C for 6 h to remove possible adsorbates. STM image shows that the sample keeps its original structure (Figure S4b).

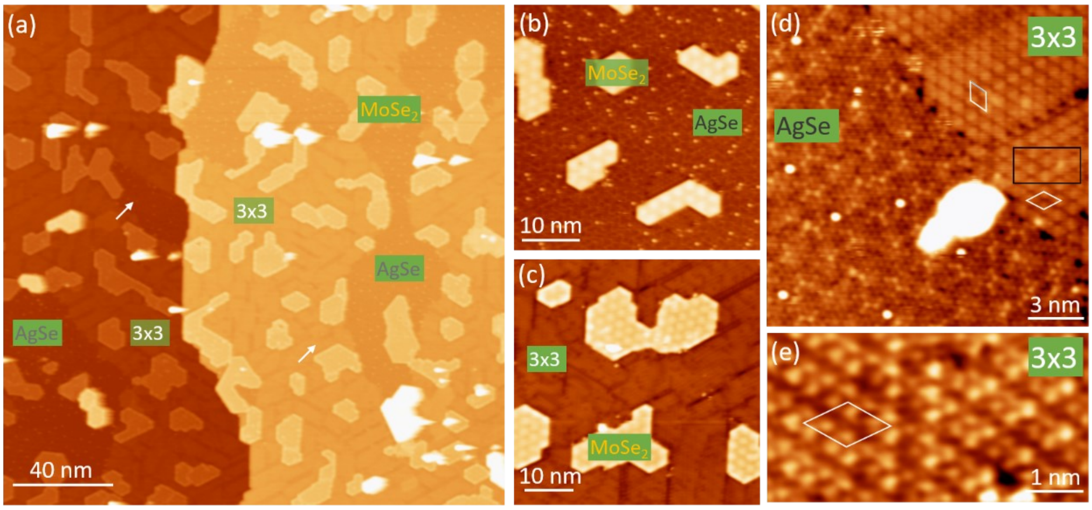

Figure S5. (a) Large-scale STM image (U_s = -3.0 V and I_t = 0.03 nA) of SL-$MoSe_2$ islands on Ag(111) substrate. (b), (c) and (d) Zoom-in STM images, showing the highly-resolved structures. (b) (U_s = -2.5 V and I_t = 0.50 nA), (c) (U_s = -2.0 V and I_t = 0.10 nA), (d) (U_s = -0.5 V and I_t = 3.0 nA). (e) Zoom-in STM image (U_s = -0.5 V and I_t = 3.0 nA) from the black rectangle in (d), showing the atomically resolved structure of 3×3 domain. The white rhombus is one of unit cell.

Figure S5a is a large-scale STM image of the as-synthesized single-layer $MoSe_2$ islands on Ag(111) substrate. Besides the $MoSe_2$ islands, there are two different apparent structures, marked by 3 × 3 and AgSe. Figure S5b and 5d are zoom-in STM images, showing the highly-resolved structures of AgSe monolayer, which is in good agreement with our previously published result[1]. Figure S5c and 5d are zoom-in STM images, showing the high-resolution structure of 3×3 superstructure. Figure S5e are the atomically resolved STM image of the 3×3 superstructure, where the white rhombus indicates one of the unit cell. The parameter of the unit cell is measured to be 0.87 nm, which is close to three times of a_{Ag} lattice. We speculate that the 3×3 superstructure may be one of silver selenide compounds.

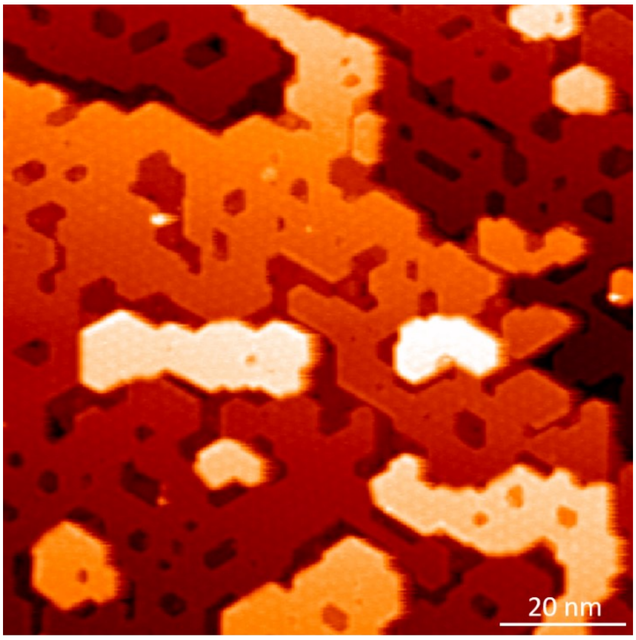

Figure S6. (a) Large-scale STM images of SL-$MoSe_2$ islands after several cycles. (U_s = -3 V and I_t = 0.02 nA).

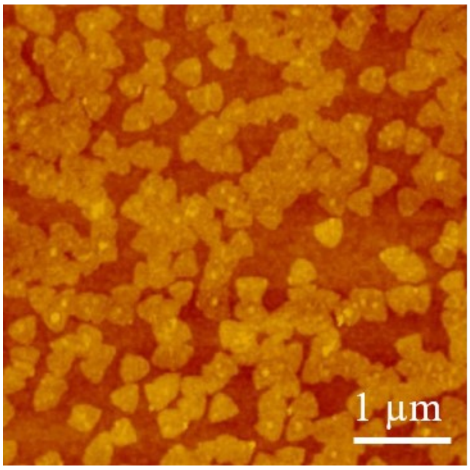

Figure S7. AFM image of CVD-synthesized SL-$MoSe_2$ on Al_2O_3 substrate. The synthetic method is reported in literatures.[3, 4]

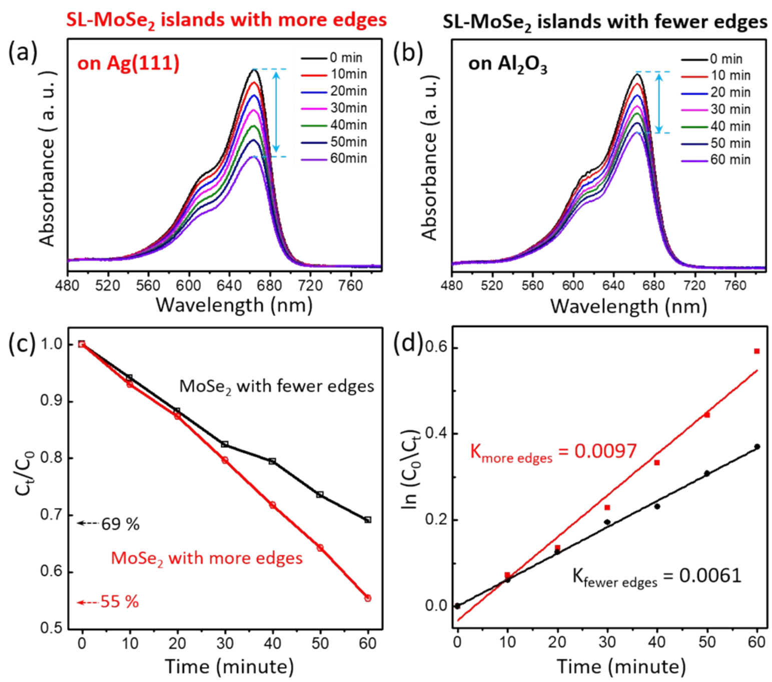

Figure S8. (a, b) photodecomposition of MB molecules by $MoSe_2$/Ag(111) with more edges (a) and $MoSe_2/Al_2O_3$ with fewer edges (b) under visible light irradiation. The blue two-headed arrows indicate the total decrease. (c) normalized concentration of MB solution vs time. C_t and C_0 are the MB concentrations at the light irradiation time t min and 0 min, respectively. (d) kinetic photodegradation curves for the MB solution.

To emphasize the importance of the number of edge sites in improving the photocatalytic activity, we performed the photocatalytic measurements for CVD synthesized SL-$MoSe_2$ on Al_2O_3 substrate. Figure S7 shows a typical AFM image of single-layer $MoSe_2$ islands with an average size of 500 nm, which is obviously larger than that of SL-$MoSe_2$ islands on the Ag(111) substrate (Figure 5). We then evaluated the photocatalytic activity of the SL-$MoSe_2$/ Al_2O_3 by the degradation of MB solution under the visible light irradiation. Figure S8(b) is the photodecomposition of MB molecules for the $MoSe_2/Al_2O_3$ sample. After 60 min irradiation only 31% of MB molecules (69% MB molecules remained unchanged) were degraded for $MoSe_2/Al_2O_3$ sample (black line, labeled by $MoSe_2$ with fewer edges), smaller than the 45% of MB molecules degradation for $MoSe_2$/Ag(111) (red line, labeled by $MoSe_2$ with more edges), as shown in Figure S8(c). Obviously, the degradation level (marked by the blue two-headed arrow) for the $MoSe_2/Al_2O_3$ is smaller than for $MoSe_2$/Ag(111) (Figure S8(a) and 8(b)). Furthermore, the kinetic photodegradation curves for $MoSe_2$/Ag(111) and $MoSe_2/Al_2O_3$ were calculated (Figure S8d). The linear relationship were revealed with the reaction rate constants of about 0.0097 min^{-1} and 0.0061 min^{-1} for $MoSe_2$/Ag(111) (red line) and $MoSe_2/Al_2O_3$ (black line), respectively, unambiguously confirming that the nanoscale SL-$MoSe_2$ islands featured with more edges has a higher photocatalytic activity.

Reference

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