

# Is charge transfer doping possible at the interfaces of monolayer VSe<sub>2</sub> with MoO<sub>3</sub> and K?

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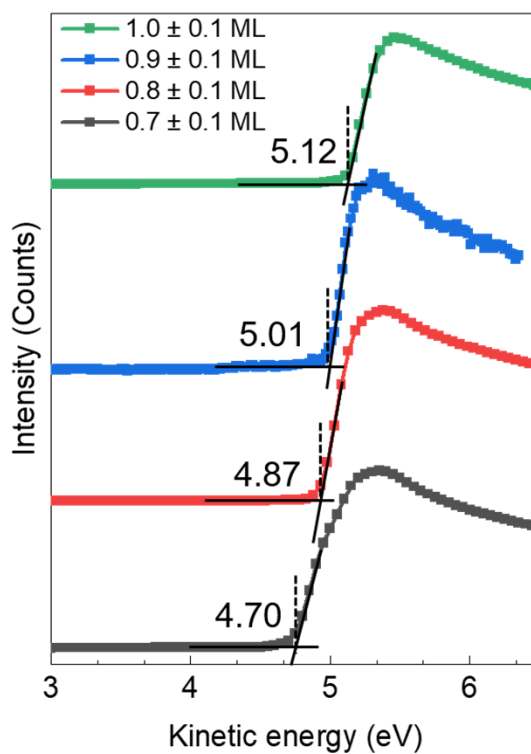
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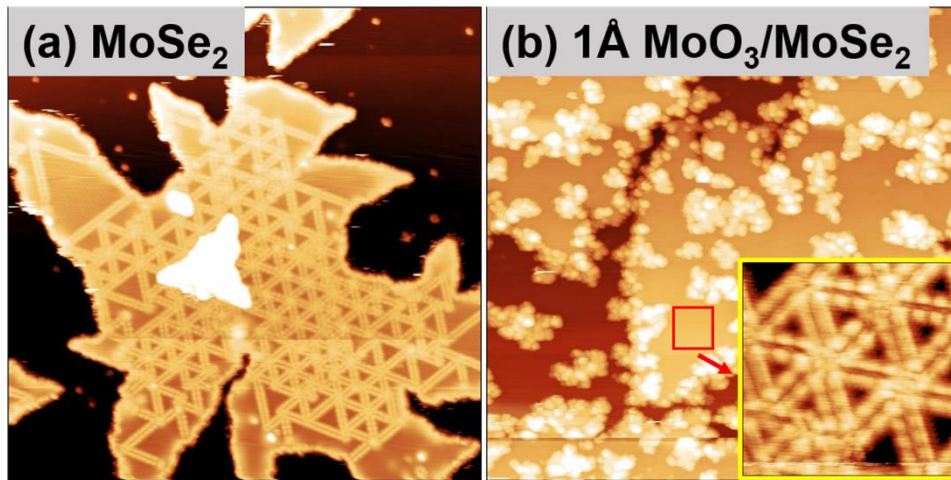
1. The work function of VSe<sub>2</sub> as a function of coverage.



**Figure S1.** The work function of VSe<sub>2</sub>/HOPG as a function of VSe<sub>2</sub> coverage. The work function increases from ~4.70 to ~5.10 eV with the increasing coverage from 0.7 ± 0.1 monolayer (ML) to 1.0 ± 0.1 ML.

## 2. The control experiment of MoO<sub>3</sub> deposition on monolayer MoSe<sub>2</sub>:

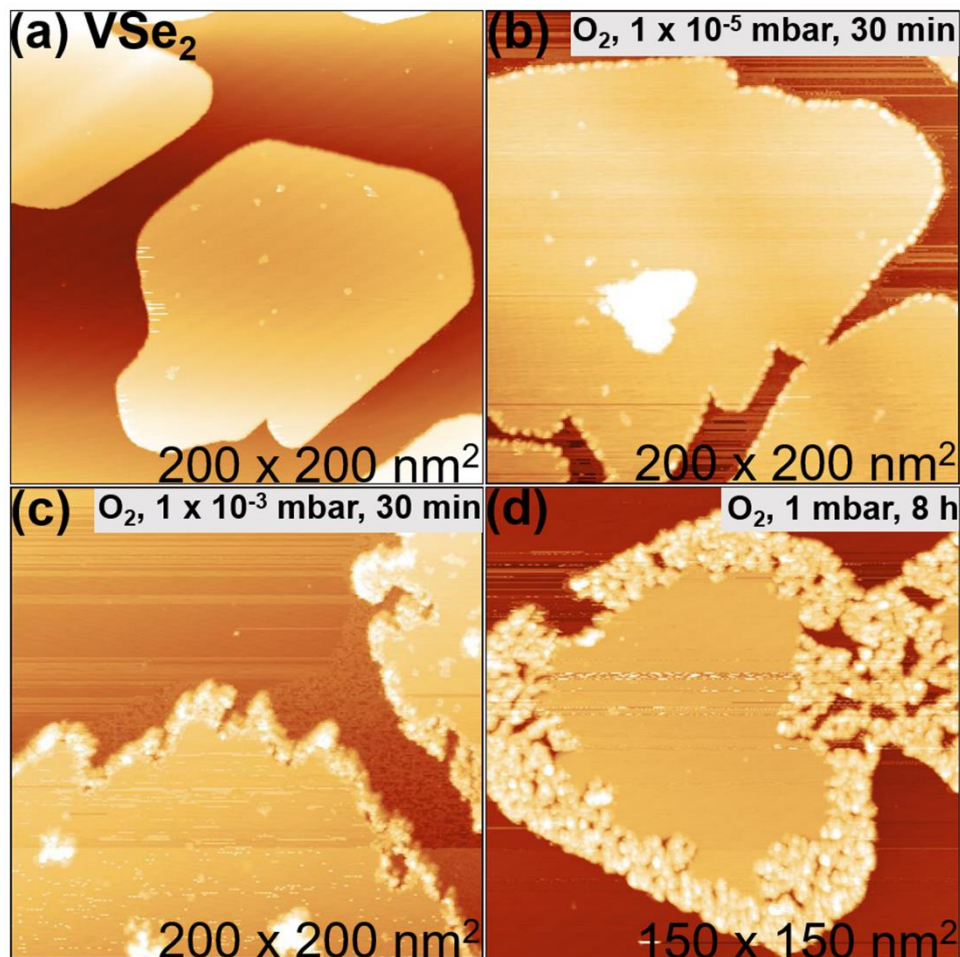
The STM of monolayer MoSe<sub>2</sub> in Figure S2a shows the dense mirror twin grain boundaries (MTGBs). Deposition of 1 Å MoO<sub>3</sub> on MoSe<sub>2</sub> is shown in Figure S2b. The inset of Figure S2b is the zoom-in image of the red square, showing the MTGBs. Although MoO<sub>3</sub> tends to adsorb on MoSe<sub>2</sub> edges, no preferential adsorption of MoO<sub>3</sub> on MTBs was observed, demonstrating that the VSe<sub>2</sub> edges are more chemically reactive than MoSe<sub>2</sub> grain boundaries.



**Figure S2.** STM images of (a) MoSe<sub>2</sub> and (b) 1 Å MoO<sub>3</sub> on MoSe<sub>2</sub>. (Scan size: 100 × 100 nm<sup>2</sup> for both (a) and (b) and 13 × 13 nm<sup>2</sup> for the inset of (b); Setpoints: a, 1.5 V, 10.0 pA; b, -2.8 V, 11.0 pA; Inset of (b), -0.7 V, 100.0 pA)

### 3. Oxygen-exposure experiment of monolayer VSe<sub>2</sub>.

The oxygen-exposure experiment of monolayer VSe<sub>2</sub> indicates that the VSe<sub>2</sub> edges are locally and easily oxidized by O<sub>2</sub>, verifying its high chemical activity.



**Figure S3.** STM images of monolayer VSe<sub>2</sub> exposed to O<sub>2</sub>. (a) Pristine VSe<sub>2</sub>; (b) – (c) VSe<sub>2</sub> exposed to an O<sub>2</sub> partial pressure of  $\sim 1 \times 10^{-5}$  mbar for 30 min,  $\sim 1 \times 10^{-3}$  mbar for 30 min and  $\sim 1$  mbar for 8 hours, respectively. (Setpoints: a, -1.4 V, 20.0 pA; b, -1.0 V, 24.0 pA; c, -1.4 V, 19.0 pA; d, -2.0 V, 21.0 pA)