## **Supporting Information**

## **Role of Chemical Potential in Flake Shape and Edge Properties of Monolayer MoS<sub>2</sub>**

Dan Cao,<sup>†</sup> Tao Shen,<sup>‡</sup> Pei Liang,<sup>‡</sup> Xiaoshuang Chen,<sup>§</sup> and Haibo Shu<sup>\*,<sup>‡</sup>,§</sup>

<sup>†</sup>College of Science, China Jiliang University, 310018 Hangzhou, China

<sup>‡</sup>College of Optical and Electronic Technology, China Jiliang University, 310018 Hangzhou, China

<sup>§</sup>National Laboratory for Infrared Physics, Shanghai Institute of Technical Physics, Chinese Academy of Science, 200083 Shanghai, China

\* Correspondence should be addressed to Haibo Shu, shu123hb@gmail.com,

The computational models of armchair and zigzag MoS<sub>2</sub> edges



**Figure S1** Atomic structures of (a) armchair (AC) and (b) zigzag (ZZ) MoS<sub>2</sub> nanoribbons.  $W_A$  and  $W_Z$  represent the width of armchair and zigzag nanoribbons respectively, and  $L_A$  and  $L_Z$  denote the length of armchair and zigzag nanoribbons along their periodic direction, respectively.

## The zero-point energies of various MoS<sub>2</sub> edges

	, 0	
Edge type	Edge structure	E <sub>ZPE</sub> (eV/Å)
Armchair	AC	0.018
Zigzag-perfect	ZZ-Mo	0.035
	ZZ-S	0.029
	ZZ-Mo2	0.052
	ZZ-S2	0.034
Zigzag-defect	ZZ-57-S	0.995
	ZZ-57-Mo	0.114

**Table S1.** Zero-point energies ( $E_{ZPE}$ ) of various MoS<sub>2</sub> edges, including of armchair (AC), perfect (ZZ-perfect) and deficient zigzag (ZZ-defect) edges.

The spin-density distribution of hexagonal MoS<sub>2</sub> cluster



Figure S2 (a) Top and (b) side views of the spin-density isosurface distribution of a hexagonal  $MoS_2$  cluster. In the cluster, three ZZ-S edges are spin-polarized and three ZZ-S2 edges are spin-unpolarized.

## The DOS of triangular MoS<sub>2</sub> cluster



Figure S3 Total DOS and local DOS of Mo and S atoms of the triangle  $MoS_2$  cluster presented in Figure 5b. The red and blue lines denote the DOS of Mo and S atoms, respectively. The Fermi level is set to energy zero.