Supporting Information

Effect of Light illumination and Se vacancy Toward Fast Oxidation of Ultrathin Gallium selenide

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1. Adsorbed structures of H_2O and O_2

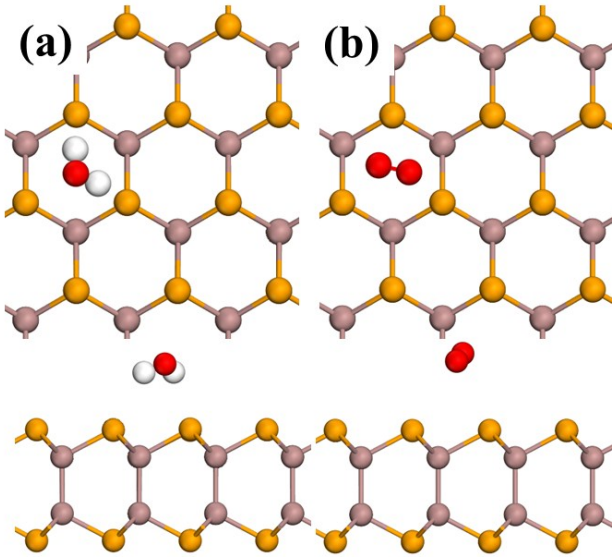

Fig. S1. Structures (top and side views) of H_2O (a) and O_2 (b) adsorb on vacancy-free GaSe surface, respectively. Ga, Se, O and H atoms are labeled as brown, yellow, red and white colors, respectively.

2. Reaction pathways of O_2 dissociation on perfect surface.

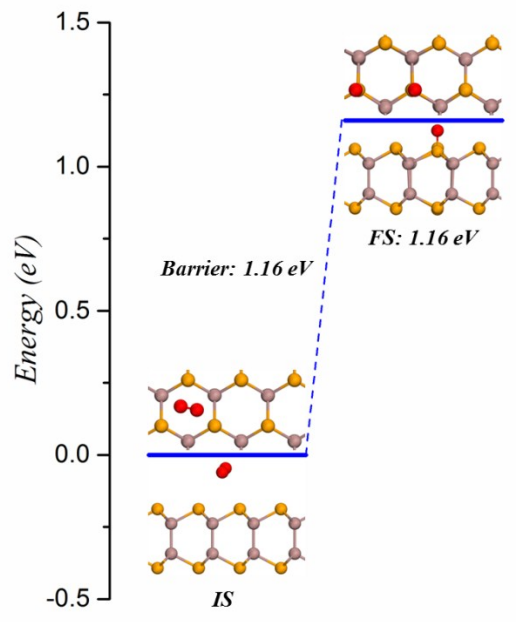

Fig. S2. Energy difference of initial (O_2 physisorbed on the surface of GaSe) and final state (O2 dissociated and adsorbed onto surface of GaSe and formed Se-O bond)

3. HSE band for monolayer GaSe from monolayer to four-layer

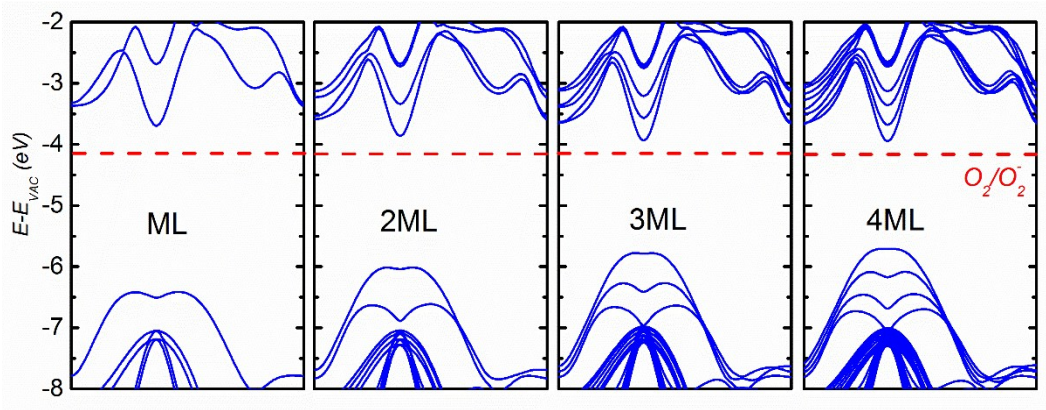

Fig. S3. The HSE band structure for monolayer to four layer of GaSe. The dashed line identifies the redox potential of O_2/ O_2^-.

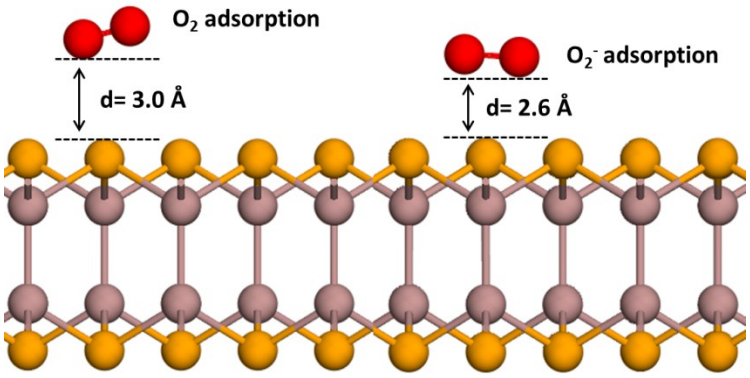

Fig. S4 The adsorption structure of O_2 and O_2^- physically adsorbing on the surface GaSe.

Table S1 Three types of attacking Ga-Se bonds and their energy.

Type of Ga-Se bond	Ⅰ Ⅳ	Ⅰ Ⅲ or Ⅱ Ⅳ	Ⅱ Ⅲ
Energy (eV)	203.594	203.261	203.261

4. Adsorbed structures of H_2O and O_2 on SeVs

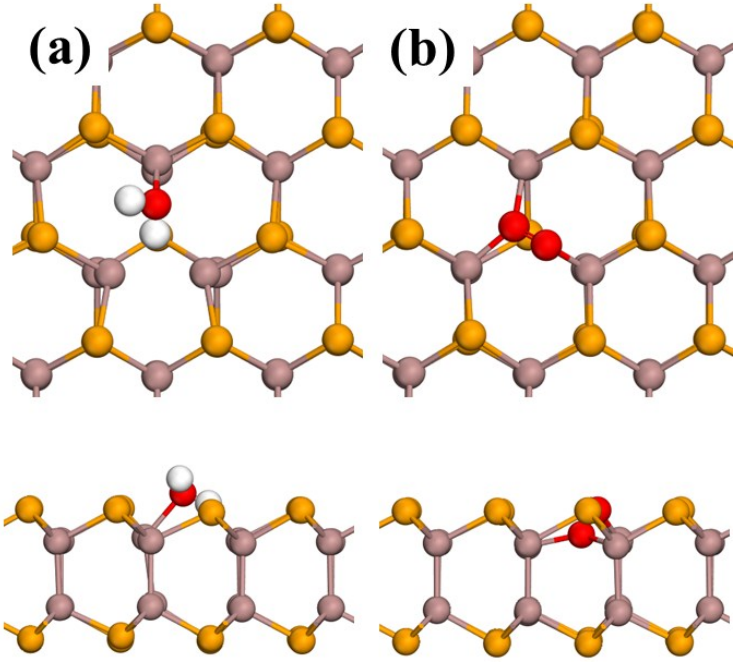

Fig. S5. Structures (top and side views) of H_2O (a) and O_2 (b) adsorb on Se-vacancy GaSe surface, respectively.

5. Different reaction products of attacking Ga-Se bond

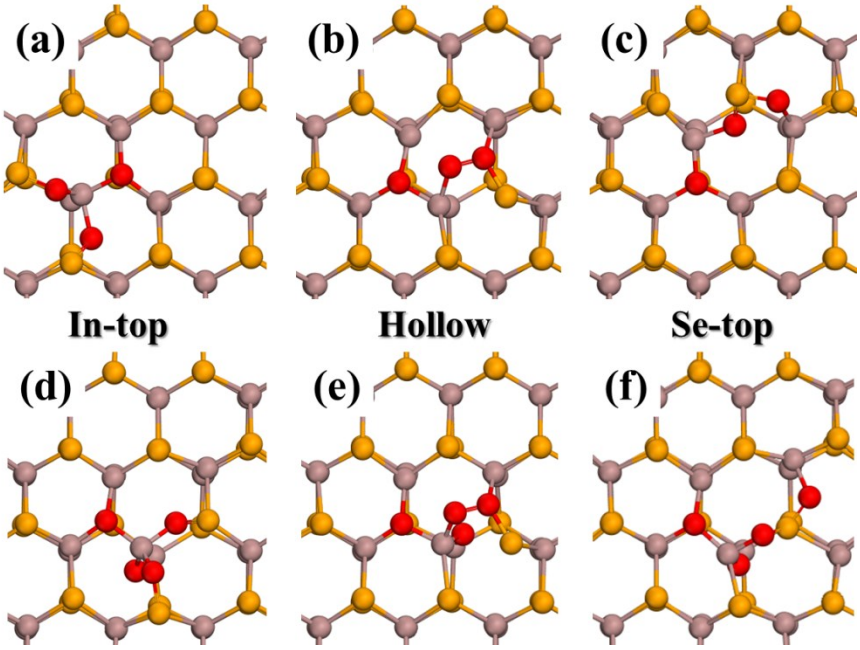

Fig. S6. Different reaction pathways and products of attacking Ga-Se bond: In-top, Se-top and hollow. (a), (b) and (c) for H_2O-attacked SeVs; (d), (e) and (f) for O_2-attacked SeVs.

Table S2 Energy difference (ΔE, eV) of three kinds of final reaction products with respect to physisorption. $\Delta E = E_{chem.} - E_{phy}$

O_2 attacks sites	In-top	Hollow	Se-top
H_2O	-1.39	0.63	-1.72
O_2	-1.86	0.40	-1.17