Supporting Information for

Two-Dimensional Germanium Monochalcogenide Photocatalyst for Water Splitting under Ultraviolet, Visible to Near-infrared Light

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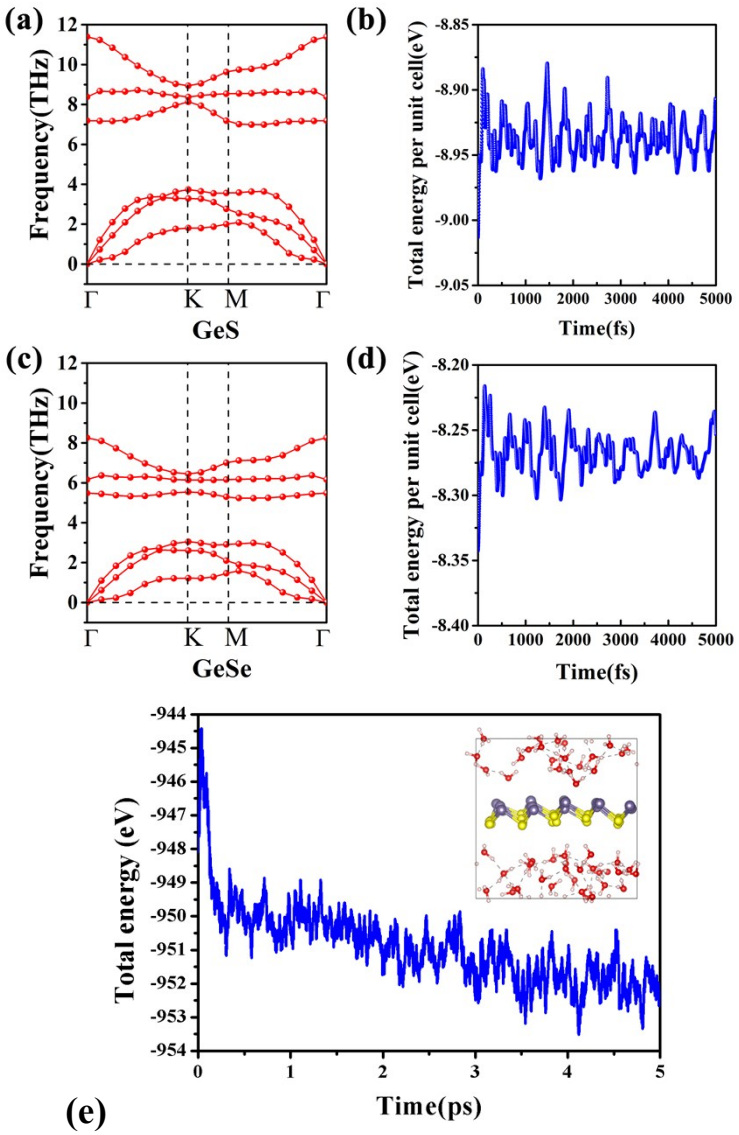

Figure S1. The phonon spectrum of monolayer (a) GeS and (c) GeSe and the vibration of total energies of (b) GeS and (d) GeSe at 300K during 5 ps by first-principles molecular dynamics calculations. (e) is the MD calculations for GeS monolayer in water environment at 300K.

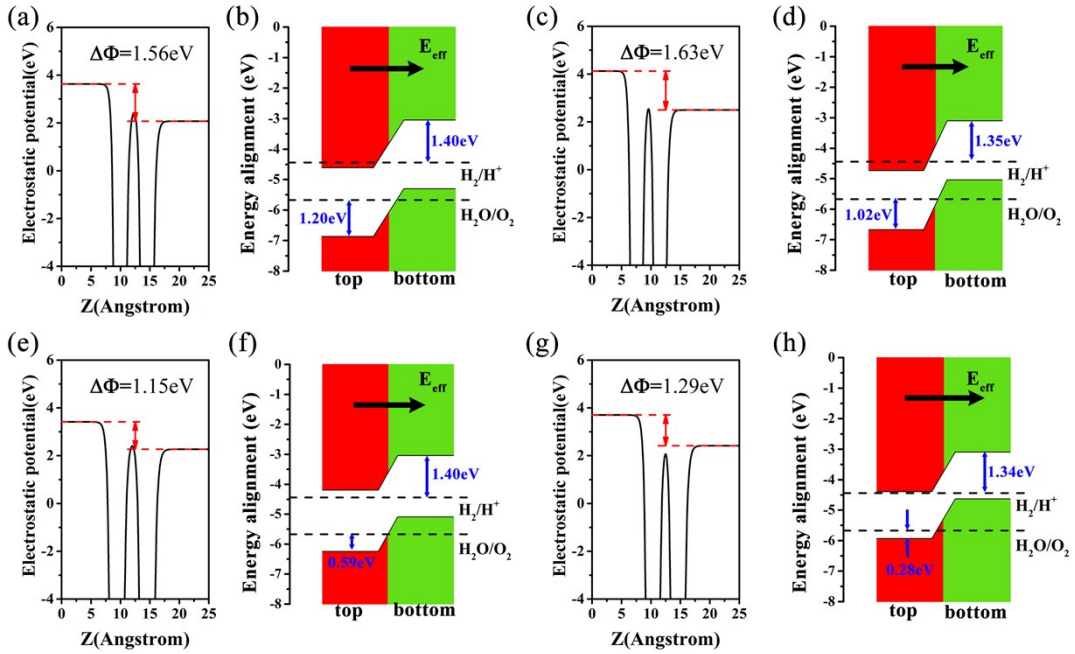

Figure S2. The electrostatic potential differences (a, c, e and g) and the energy alignments (b, d, f and g) of bilayer GeS with (a, b) AB stacking, (c, d) AC stacking, bilayer GeSe with (e, f) AB stacking and (g, h) AC stacking.

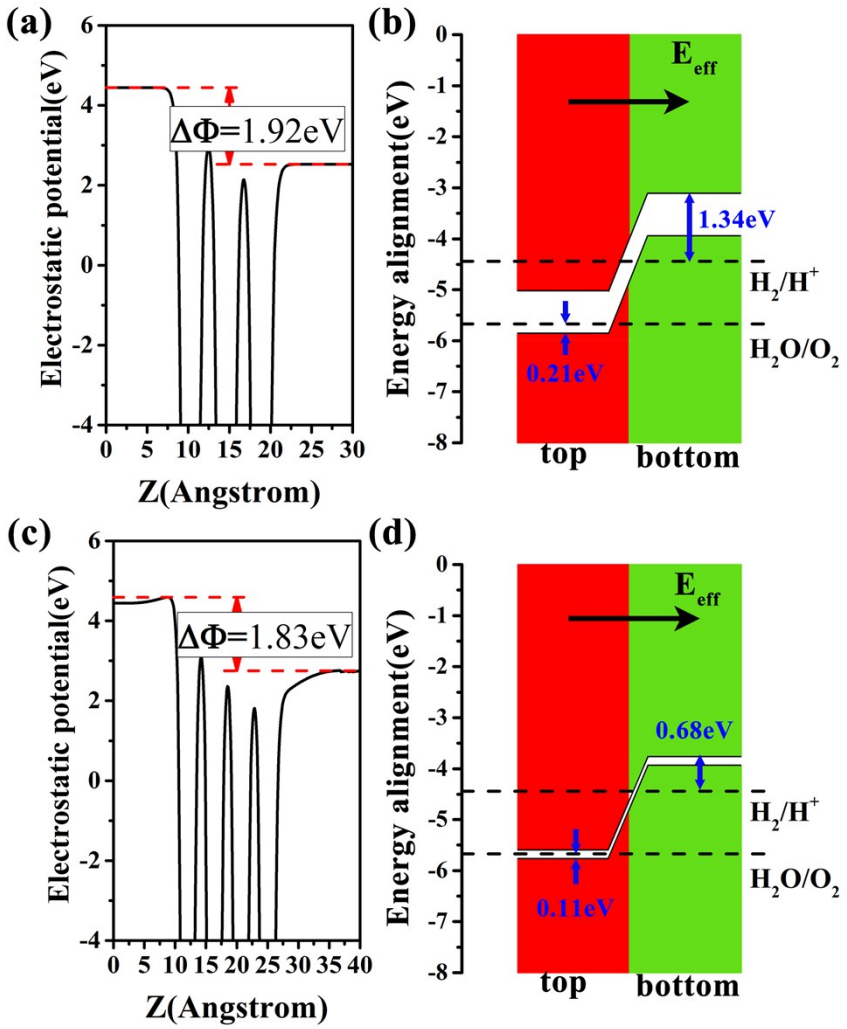

Figure S3. The surface potential differences and alignments of energy levels for (a, b) trilayer GeSe and (c, d) four-layer GeSe.

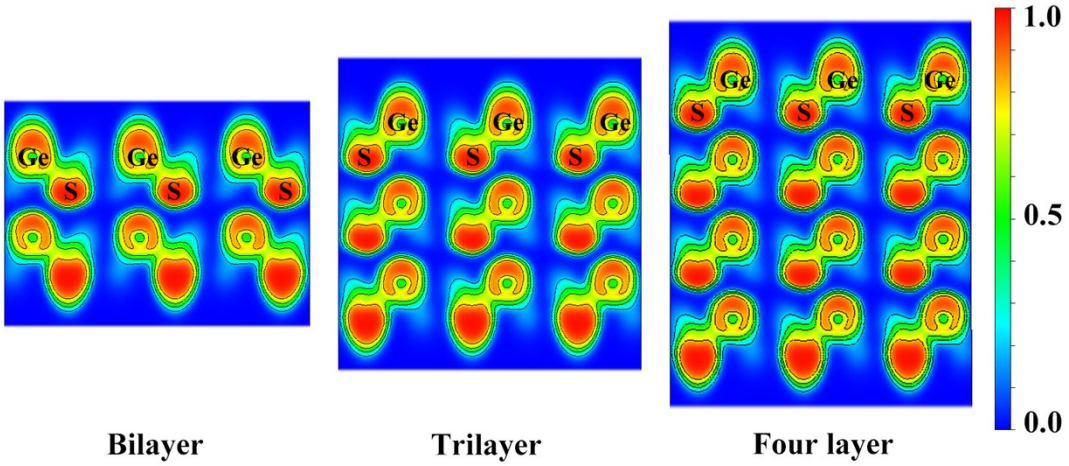

Figure S4. The electron localization function (ELF) for bi-layer, tri-layer and four-layer GeS.

Table S1. Electrostatic potential difference (eV) between Sn surface and S (Se) surface for SnS and SnSe from monolayer to trilayer. ΔE_1 is the energy difference between VBM and water oxidation potential, and ΔE_2 is the energy difference between CBM level and water reduction potential.

	Layer number	Electrostatic potential difference (eV)	ΔE_1 (eV)	ΔE_2 (eV)	Band gap (eV)
SnS	monolayer	1.19	1.47	1.51	3.02
	bilayer	2.34	1.19	1.44	1.52
	trilayer	3.57	1.14	1.47	0.27
SnSe	monolayer	0.95	0.98	1.62	2.88
	bilayer	1.93	0.60	1.53	1.43
	trilayer	2.50	0.40	1.21	0.34