Supporting Information:

Two-Dimensional Gold Sulfide Monolayers with Direct Band Gap and Ultrahigh Electron Mobility

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Table S1. Lattice constants for Au_2S and AuS monolayers with or without DFT-D3 scheme included. It is clearly seen that DFT-D3 scheme has strong effects on the lattice structures. Although the gold sulfide monolayers are not layered structures, long distance dispersions cannot be ignored particularly in loose structures like γ-AuS.

2D system	α-Au_2S	β-Au_2S	α-AuS	β-AuS	γ-AuS
Lattice constants with DFT-D3	$a = b = 5.74$ Å	$a = b = 7.92$ Å	$a = 3.54$ Å $b = 6.18$ Å	$a = 6.08$ Å $b = 6.68$ Å	$a = 8.18$ Å $b = 7.16$ Å
Lattice constants without DFT-D3	$a = b = 5.78$ Å	$a = b = 8.00$ Å	$a = 3.54$ Å $b = 6.26$ Å	$a = 6.18$ Å $b = 6.91$ Å	$a = 8.77$ Å $b = 6.65$ Å

Table S2. Lattice parameters and atomic fractional positions for Au_2S and AuS monolayers.

System	Lattice parameters	Fractional coordinates
α-Au_2S	$a = b = 5.74$ Å $c = 20.0$ Å, $\theta = 90°$	Au1 (0.250, 0.750, 0.453), Au2 (0.250, 0.250, 0.408), Au3 (0.750, 0.750, 0.408), Au4 (0.750, 0.250, 0.408), S1 (0.500, 0.000, 0.473), S2 (0.000, 0.500, 0.342)
β-Au_2S	$a = b = 7.92$ Å $c = 20.0$ Å, $\theta = 90°$	Au1 (0.002, 0.243, 0.528), Au2 (0.990, 0.743, 0.403), Au3 (0.246, 0.999, 0.403), Au4 (0.746, 0.987, 0.528), Au5 (0.490, 0.243, 0.528), Au6 (0.502, 0.743, 0.403), Au7 (0.746, 0.499, 0.528), Au8 (0.246, 0.487, 0.403), S1 (0.246, 0.743, 0.338), S2 (0.746, 0.243, 0.593), S3 (0.746, 0.743, 0.466), S4 (0.246, 0.243, 0.466)
α-AuS	$a = 3.54$Å, $b = 6.18$ Å $c = 20.0$ Å, $\theta = 90°$	Au1 (0.500, 0.500, 0.359), Au2 (0.000, 0.000, 0.359), S1 (0.500, 0.822, 0.301), S2 (0.500, 0.179, 0.418)
β-AuS	$a = 6.08$ Å, $b = 6.68$ Å $c = 20.0$ Å, $\theta = 90°$	Au1 (0.250, 0.250, 0.477), Au2 (0.750, 0.750, 0.477), Au3 (0.750, 0.250, 0.477), Au4 (0.250, 0.750, 0.477), S1 (0.320, 0.500, 0.399), S2 (0.680, 0.500, 0.399), S3 (0.820, 0.000, 0.555), S4 (0.180, 0.000, 0.555)
γ-AuS	$a = 8.18$ Å, $b = 7.16$ Å $c = 20.0$ Å, $\theta = 90°$	Au1 (0.249, 0.250, 0.385), Au2 (0.249, 0.750, 0.385), Au3 (0.749, 0.750, 0.385), Au4 (0.749, 0.250, 0.385), S1 (0.402, 0.000, 0.422), S2 (0.096, 0.500, 0.348), S3 (0.902, 0.500, 0.422), S4 (0.595, 0.000, 0.348)

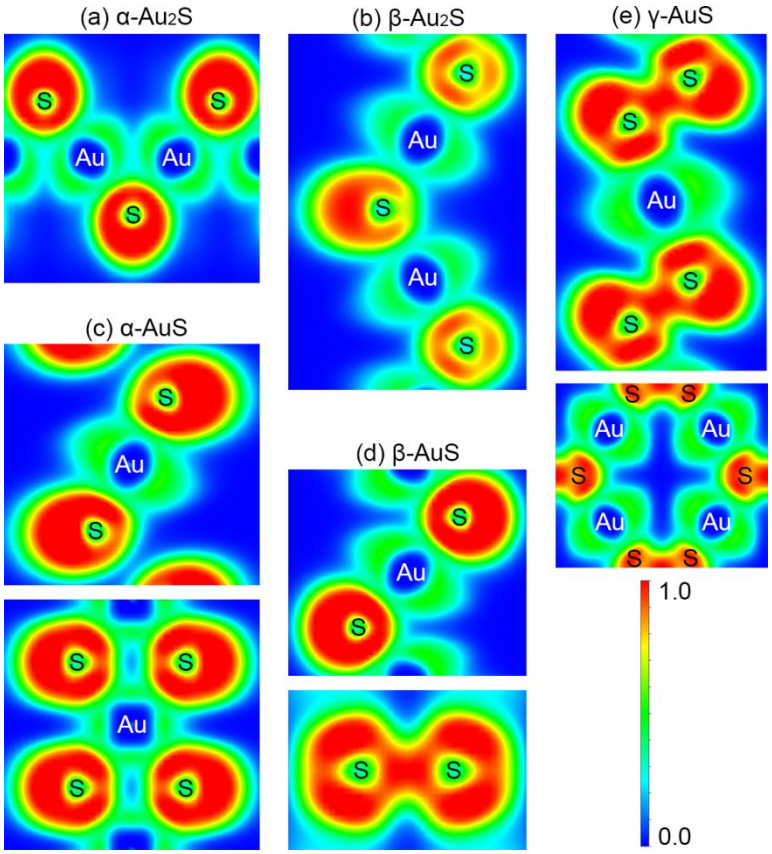

Figure S1. Electron localization functions (ELF) for Au_2S and AuS monolayers.

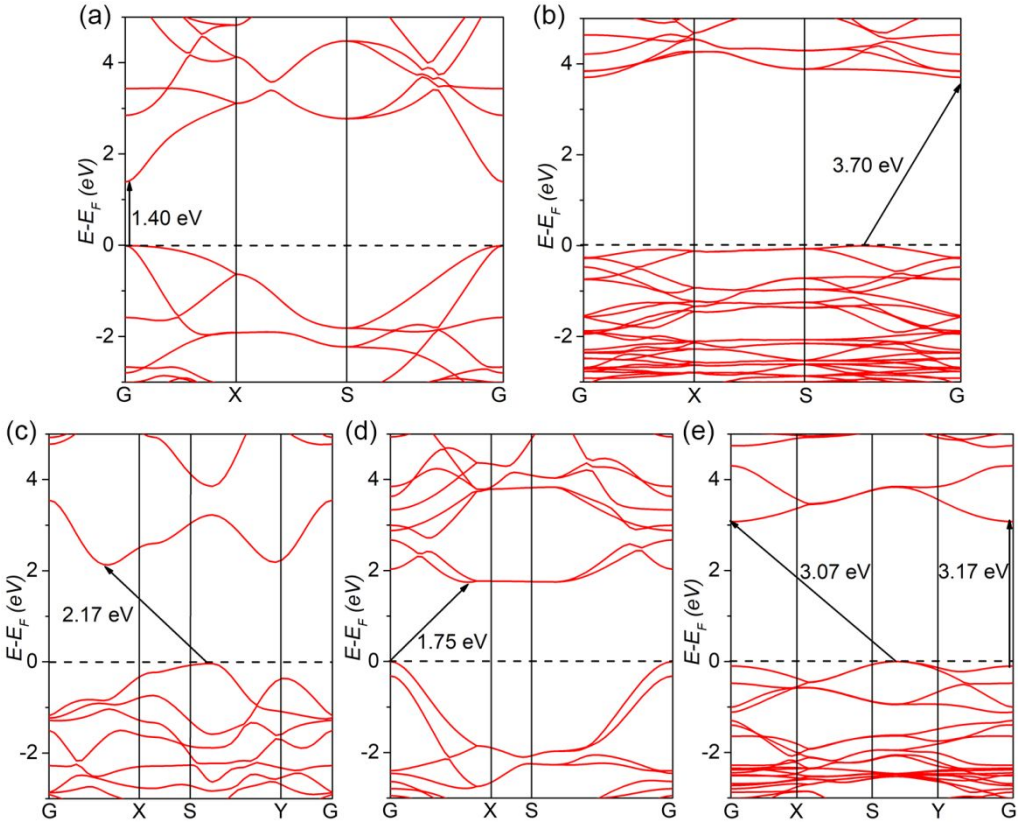

Figure S2. Band structures calculated by HSE06 functional without including spin-orbital coupling (SOC) effects for (a) α-Au_2S, (b) β-Au_2S, (c) α-AuS, (d) β-AuS and (e) γ-AuS monolayers. It is clearly seen that, to some degree, SOC reduces the band gaps of the Au_2S and AuS monolayers, as existing in many other 2D materials with heavy atoms.[1]

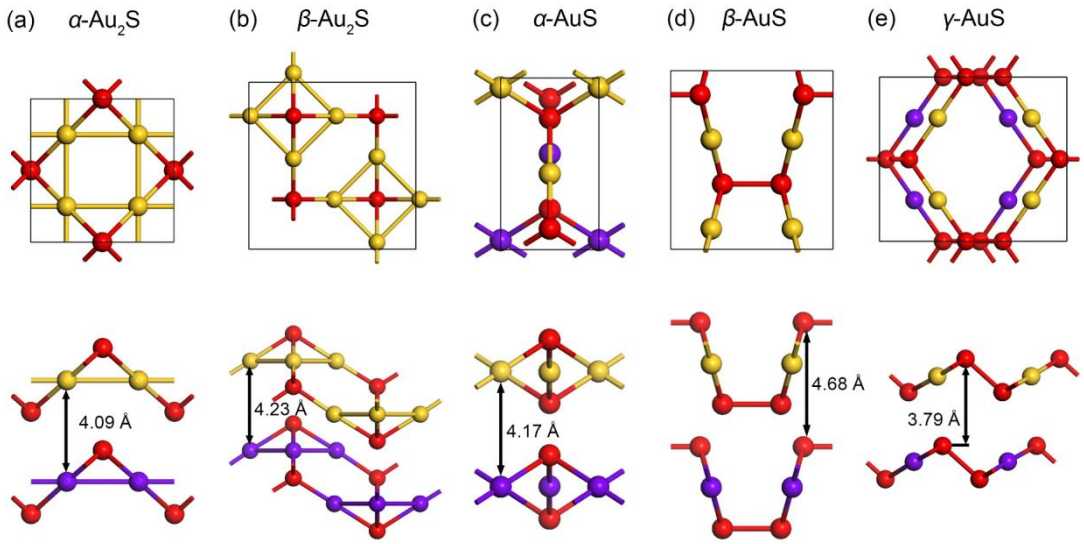

Figure S3. Atomic geometries (top and side views) of bilayer systems for 2D Au_2S and AuS structures with their layer distances indicated. Gold and violet atoms denote Au atoms in two layers, respectively. Red atoms stand for S atoms.

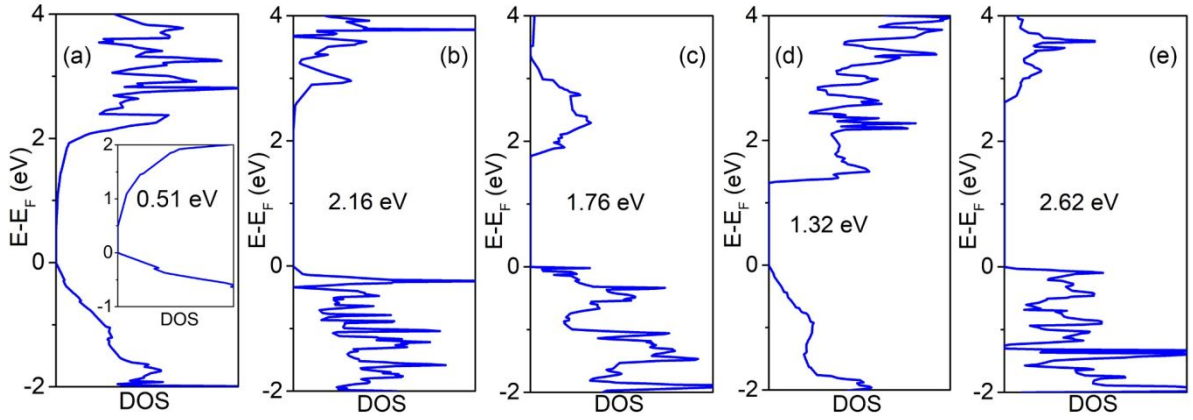

Figure S4. Density of states (DOS) of bilayer systems for 2D (a) α-Au_2S, (b) β-Au_2S, (c) α-AuS, (d) β-AuS and (e) γ-AuS structures calculated on basis of HSE06 functional with SOC effects included.

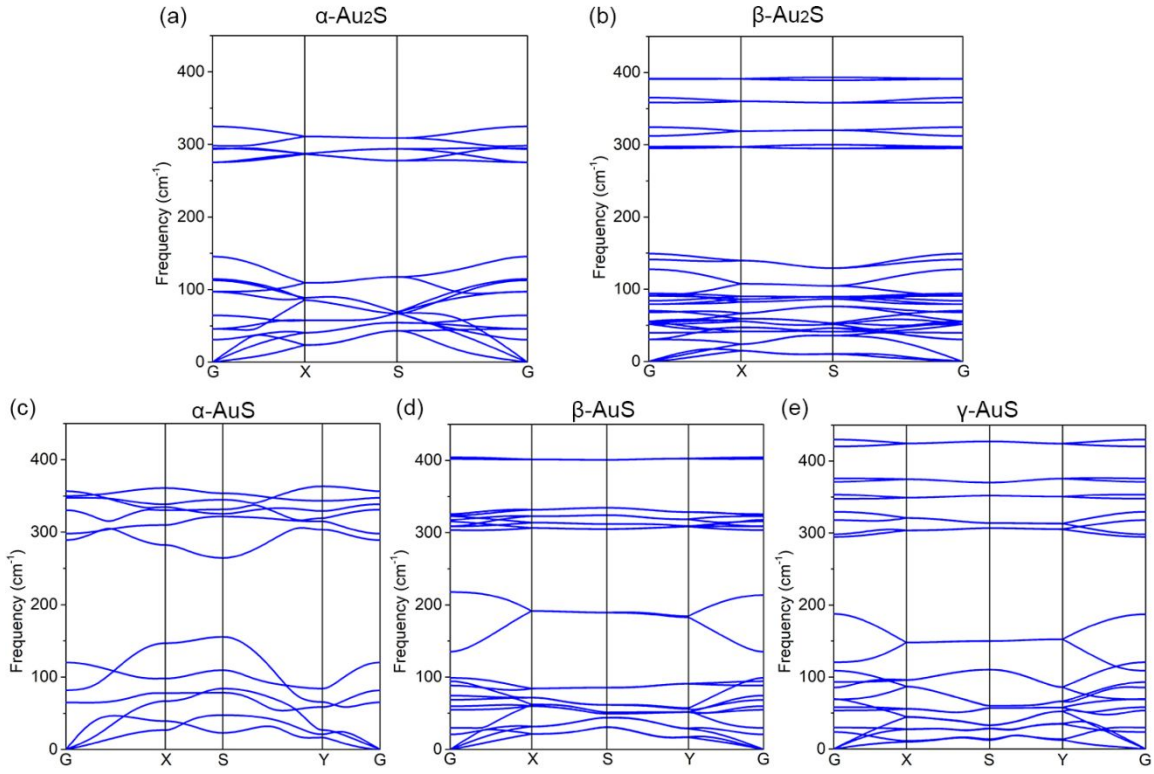

Figure S5. Phonon spectrums for α-Au_2S (a), β-Au_2S (b), α-AuS (c), β-AuS (d) and γ-AuS (e) monolayers.

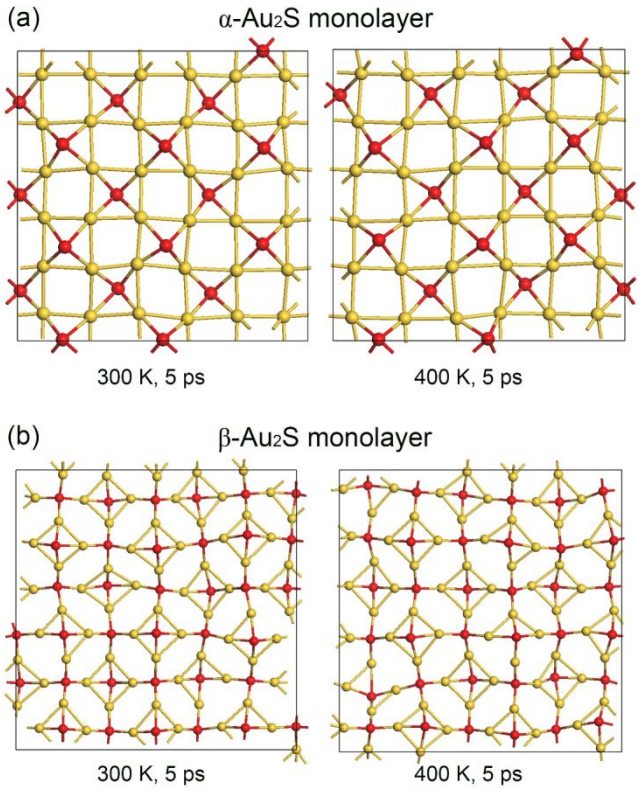

Figure S6. *Ab initio* molecular dynamics (AIMD) snapshots of Au_2S monolayers after annealing at 300 K and 400 K for 5 ps, respectively.

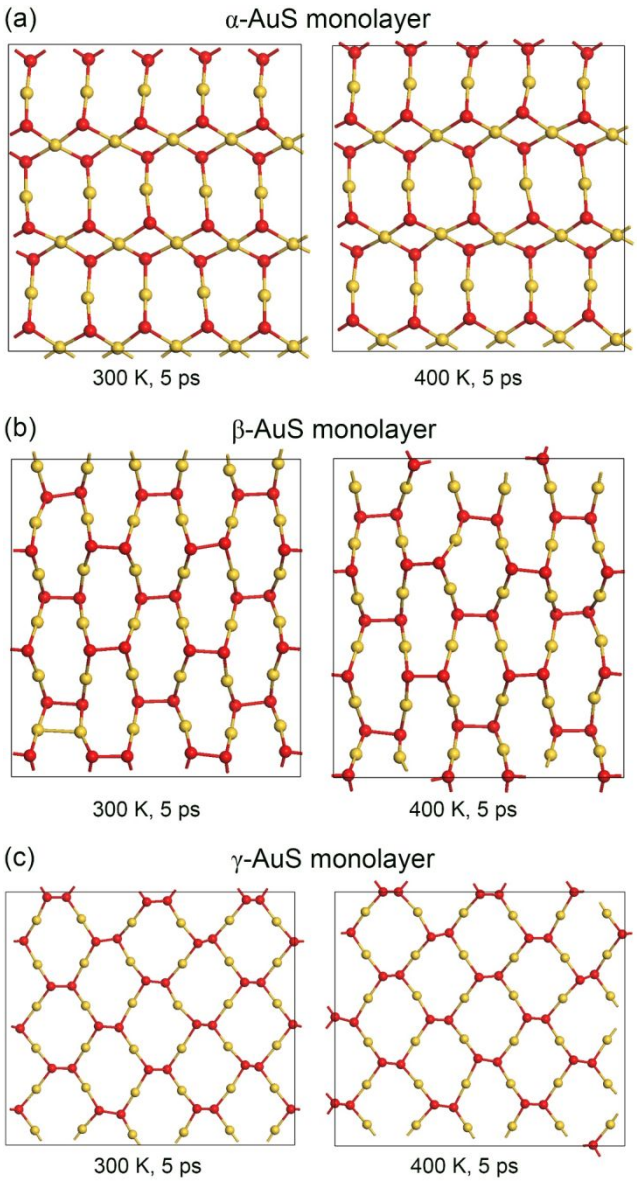

Figure S7. *Ab initio* molecular dynamics (AIMD) snapshots of AuS monolayers after annealing at 300 K and 400 K for 5 ps, respectively.

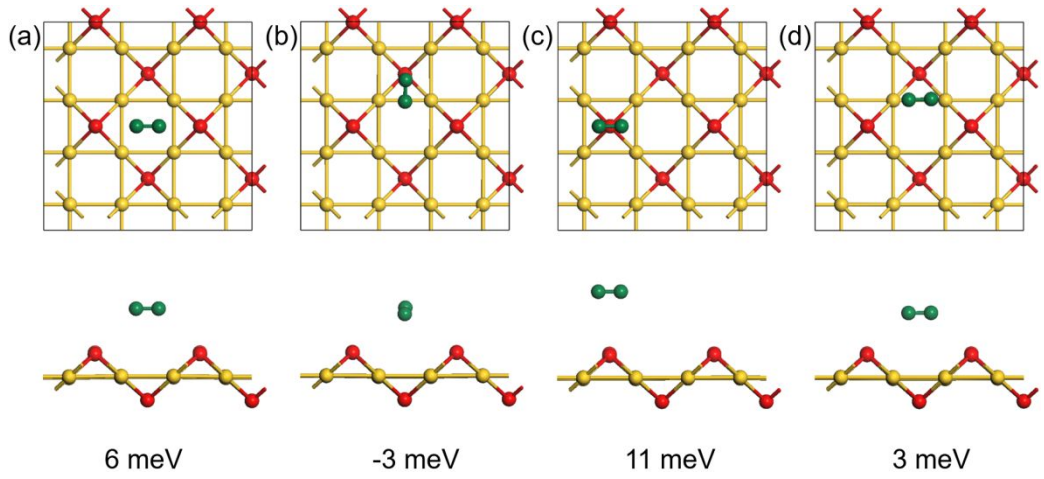

Figure S8. Top and side views for geometric structures of O_2 molecule adsorbed on α-Au_2S monolayer with adsorption energies indicated. The adsorption energies are defined as: $E_{ads} = E_{total} - E_{O_2} - E_{\alpha - Au_2S}$, where the E_{total}, E_{O_2} and $E_{\alpha - Au_2S}$ refer to energies of the system, O_2 molecule and the α-Au_2S monolayer, respectively. Negative values indicate that it is favorable for O_2 molecule to adsorb on α-Au_2S monolayer. The most stable geometry (b) is chosen for CI-NEB calculation. Gold, red and green spheres denote Au, S and O atoms, respectively.

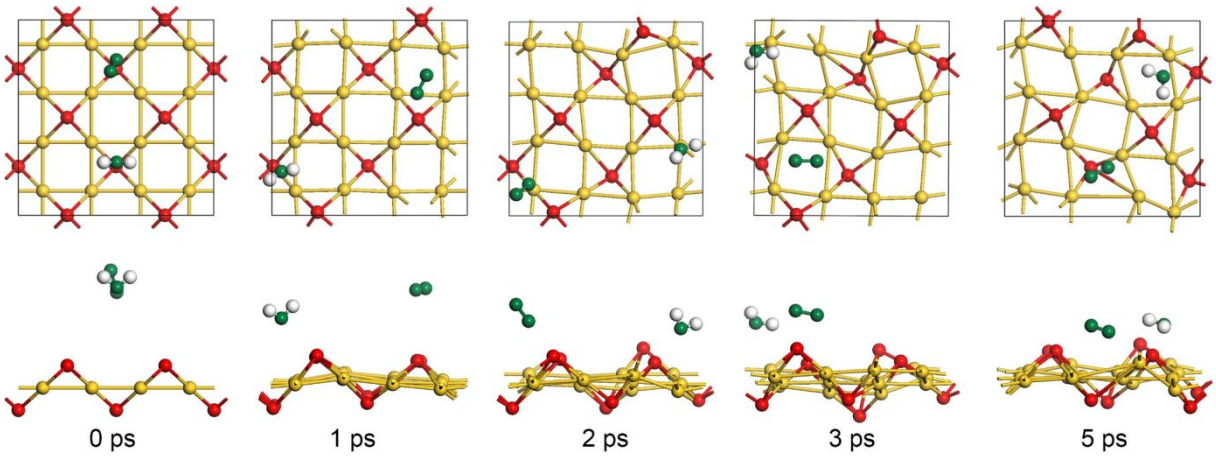

Figure S9. Snapshots of the simulated oxidation taken at 0 ps, 1ps, 2ps, 3ps and 5ps for AIMD simulations of the α-Au_2S monolayer with one O_2 and one H_2O molecule adsorbed. Gold, red, green and white spheres denote Au, S, O and H atoms, respectively.

References:

(1) Ma, L.; Dai, J.; Zeng, X. C. Two-Dimensional Single-Layer Organic-Inorganic Hybrid Perovskite Semiconductors. *Adv. Energy Mater.* **2017,** *7*, 1601731.