Supplementary Information for

Bi$_2$OS$_2$: a Direct-Gap Two-dimensional Semiconductor with High

Carrier Mobility and Surface Electron States

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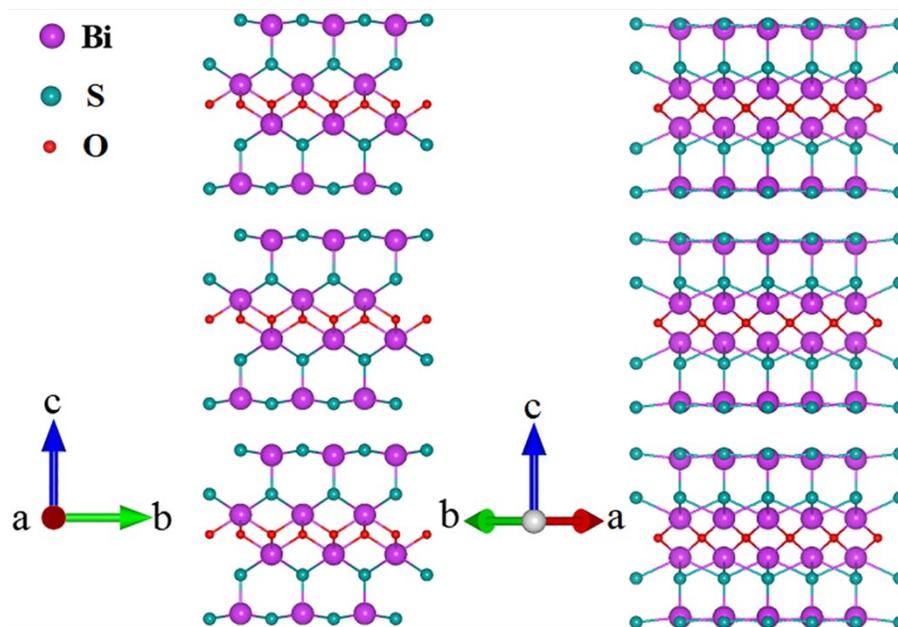
Fig. S1 Structure of bulk Bi_2OS_2 in a 3×3×3 supercell from different views.
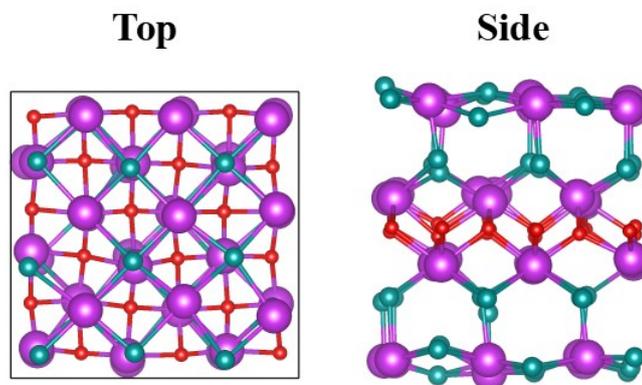
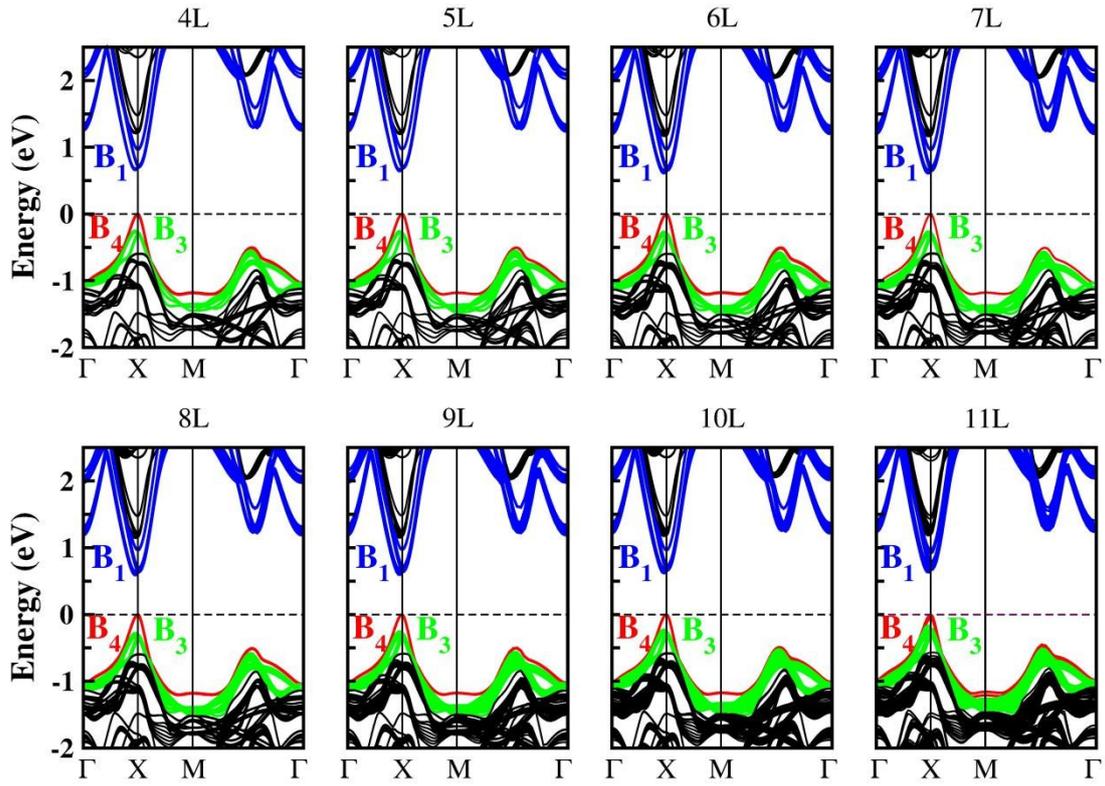**Fig. S2** Snapshots of atomic configurations of Bi_2OS_2 monolayer at temperatures of 500 K at the end of 5 ps.

Fig. S3 Calculated band structures of 2D Bi_2OS_2 from 4L to 11L. Top of the valence band is set to zero.

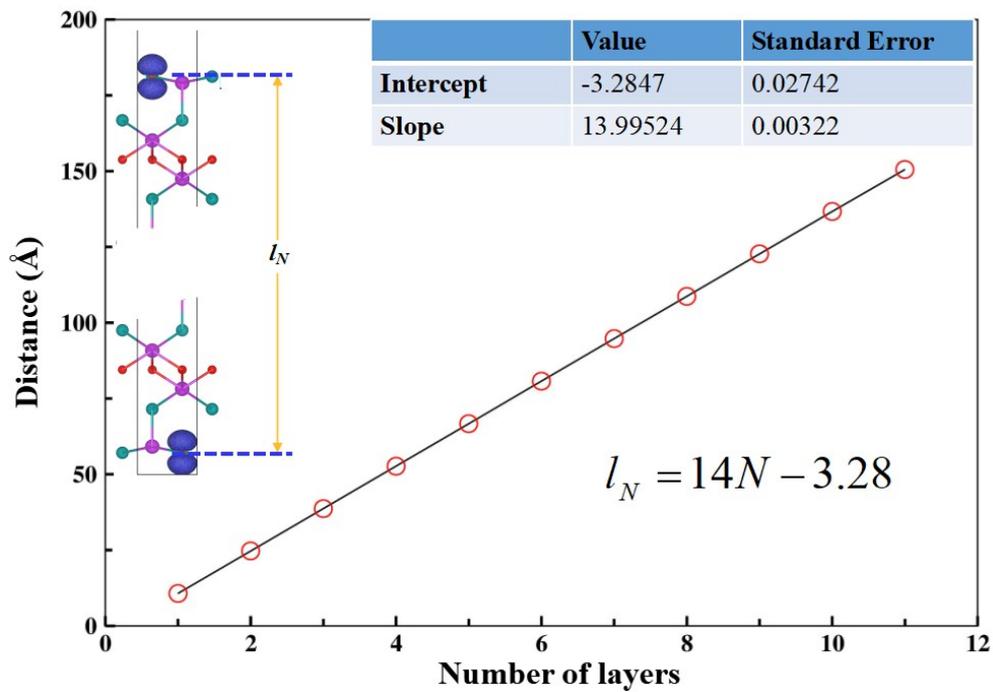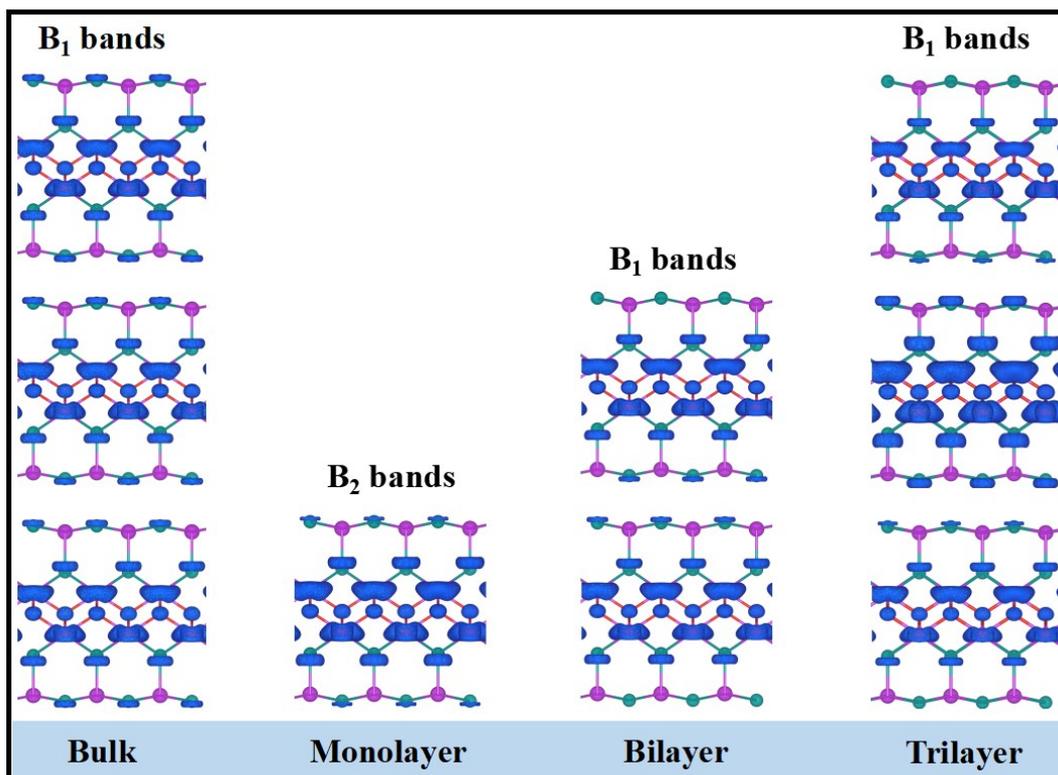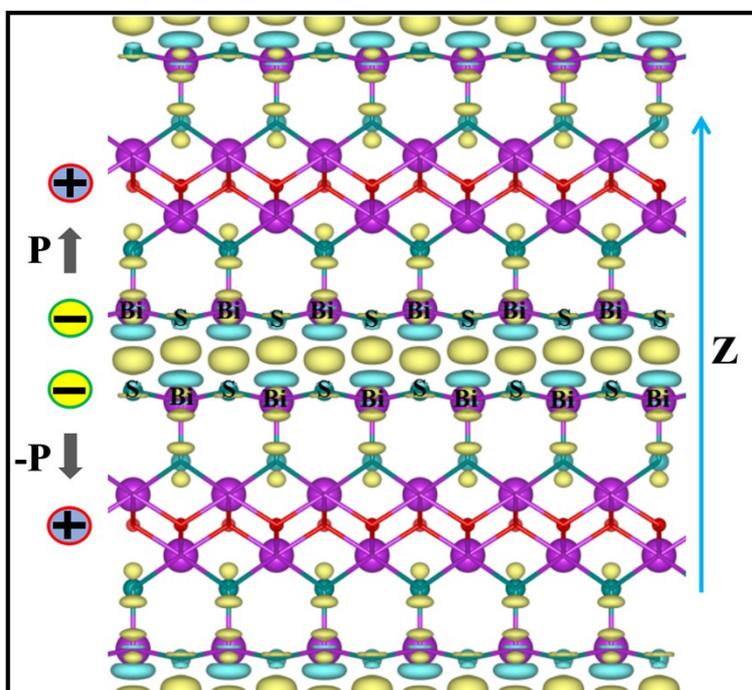|	Value	Standard Error
Intercept	-3.2847	0.02742
Slope	13.99524	0.00322

$$l_N = 14N - 3.28$$

**Fig. S4** The distance between the two outermost layers exhibits a linear dependence on the number of layers.

Fig. S5 Calculated band decomposed charge density of the B_1 and B_2 bands (labeled in Figure 2a) with the isosurface values of 0.01 $e/bohr^3$.

**Fig. S6** Different charge densities for the bulk Bi_2OS_2 with an isovalue of 0.0007 $e/bohr^3$. The cyan and yellow regions represent the charge depletion and accumulation in the space, respectively.
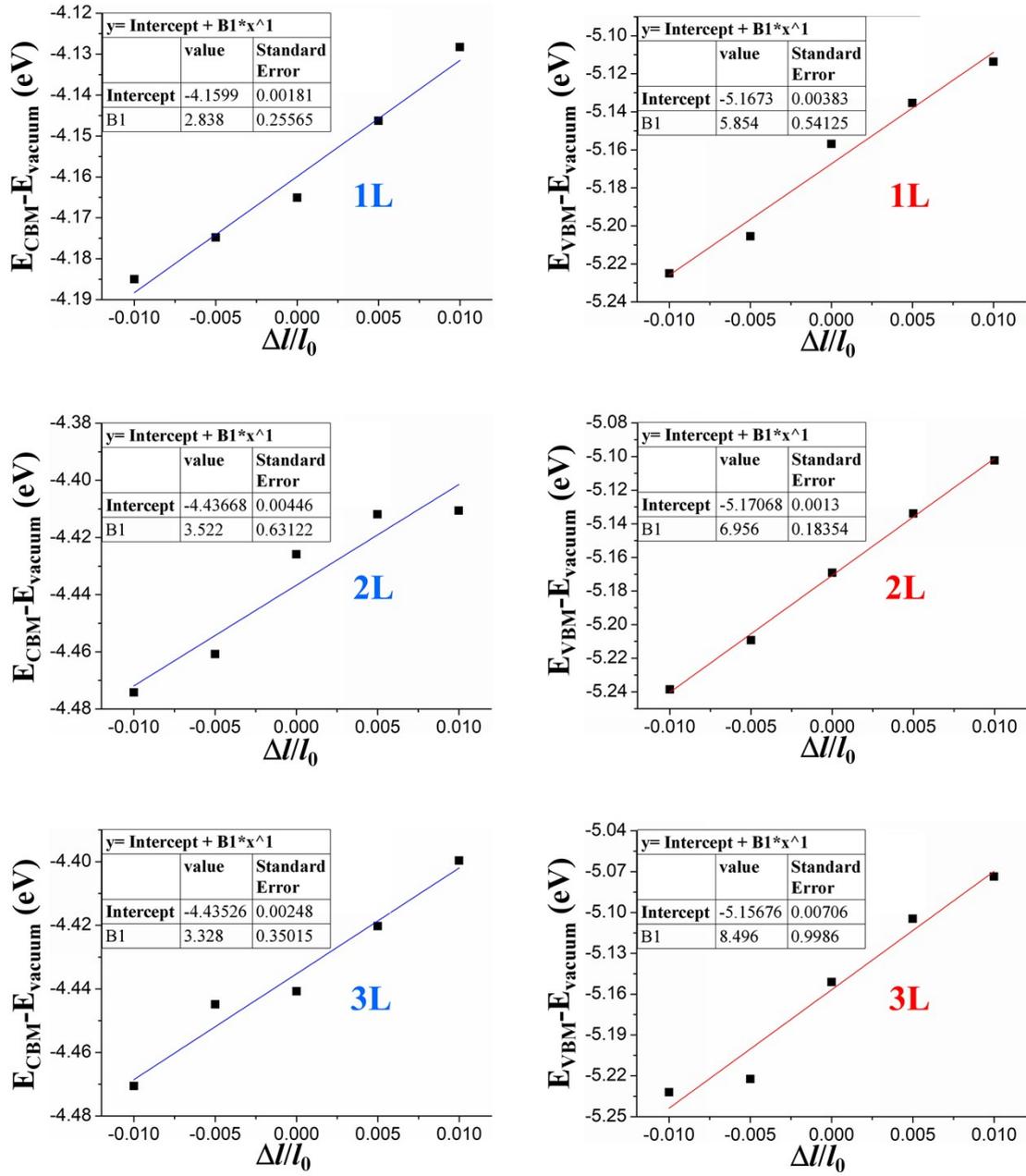
Fig. S7 Linear fitting of deformation potential. Energy change of the CBMs and VBMs of Bi_2OS_2 mono- and few layers with respect to the vacuum level as a function of lattice dilation.
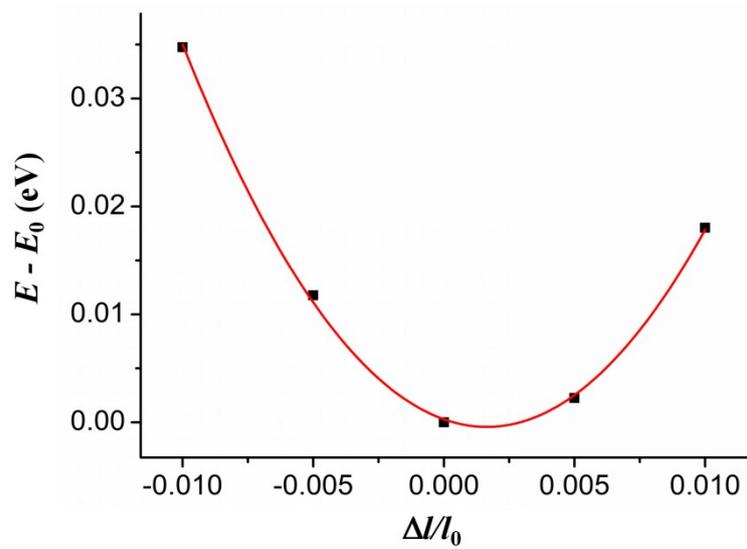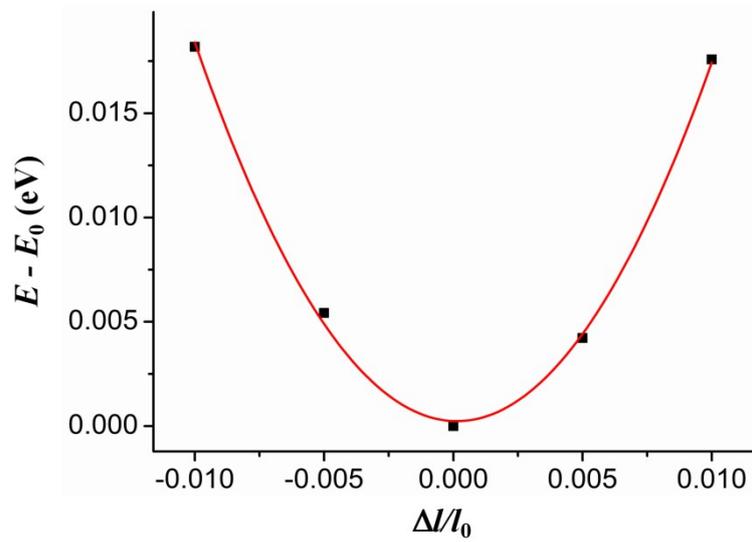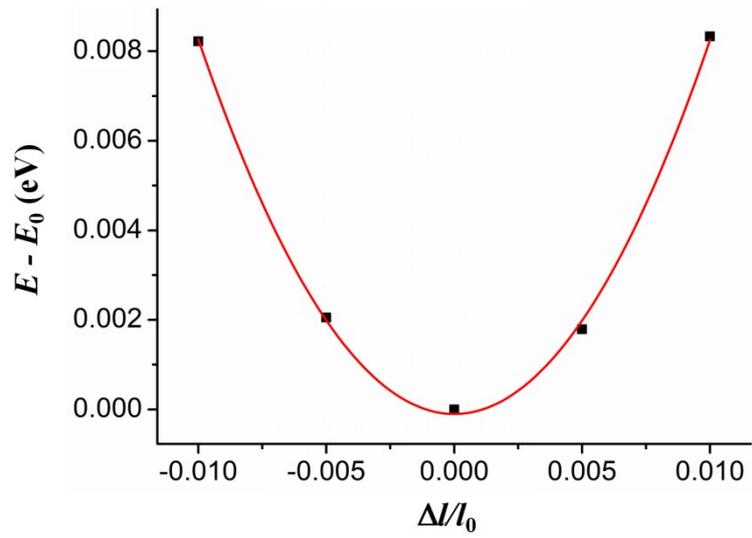
Fig. S8 Total energy-strain curve of Bi_2OS_2 mono- and few layers along x/y directions.
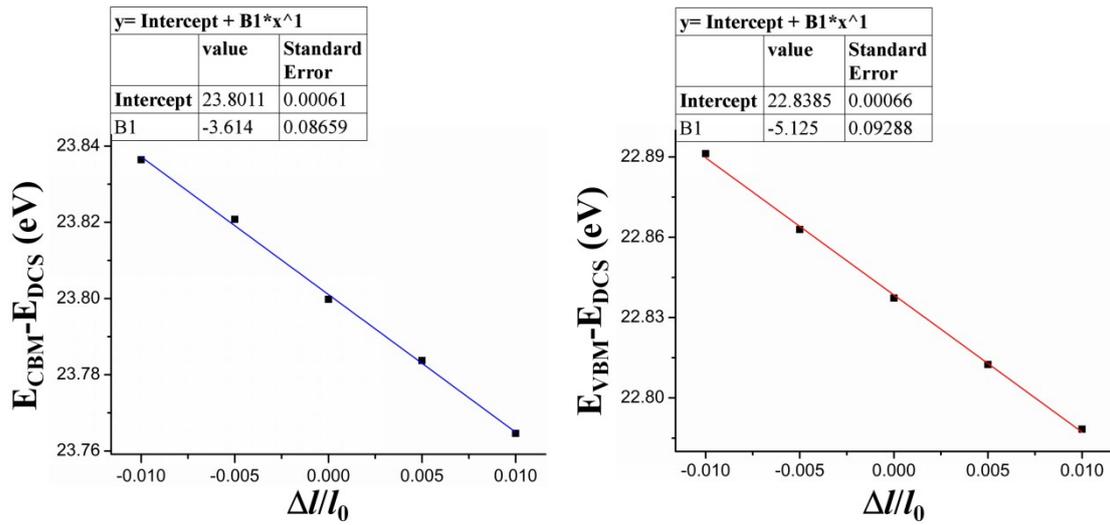
Fig. S9 Linear fitting of deformation potential. Energy change of the CBM and VBM of Bi$_2$OS$_2$ bulk with respect to the deep core states (DCS) as a function of lattice dilation.
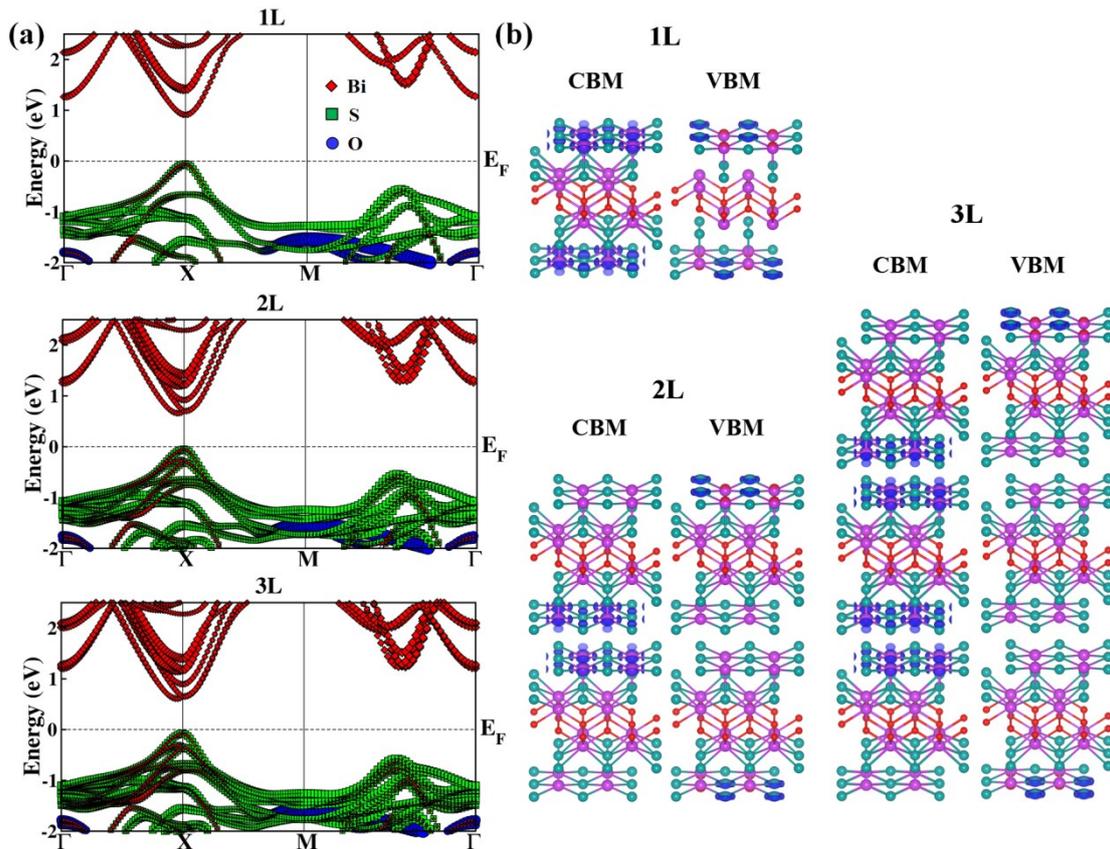
Fig. S10 (a) Atom-resolved electronic band structures of Bi$_2$OS$_2$ mono- and few-layers. The red, green, and blue dots represent the contributions from Bi, S, and O atoms. The symbol size is proportional to its population in corresponding atoms. (b)

Partial charge density is indicated in blue for conduction bands near the CBMs with an isovalue of 0.0022 e/bohr^3, and for valence bands near the VBMs with an isovalue of 0.0003 e/bohr^3. Magenta, blue-green, and red balls denote Bi, S, and O atoms, respectively.
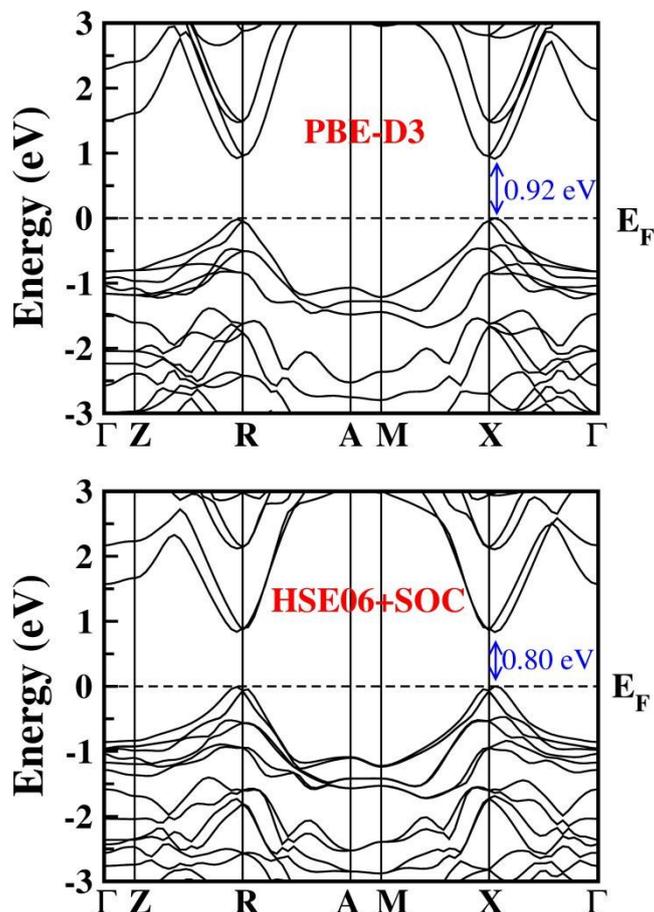
Fig. S11 Electronic band structures of bulk Bi_2OS_2 calculated at the PBE-D3 and HSE06+SOC level.

Table S1 Computed effective mass ($\textbf{\textit{m}}$*), 3D elastic modulus ($\textbf{\textit{C}}_{\textbf{3D}}$), deformation potential constant ($|\textbf{\textit{E}}^i|$), and in-plane hole and electron mobility ($\textbf{\textit{μ}}$) of Bi_2OS_2 bulk at 300 K

	Carrier Type	m*/m_e	C_{3D} (Gpa)	$|E^i|$ (eV)	$μ$ (10^3 cm^{2}V^{-1}s^{-1})
Bulk	Electron	0.23	135	3.61	28.8
	Hole	- 0.34	135	5.13	4.62