Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Tl_2S: Metal-Shrouded Two-Dimensional Semiconductor

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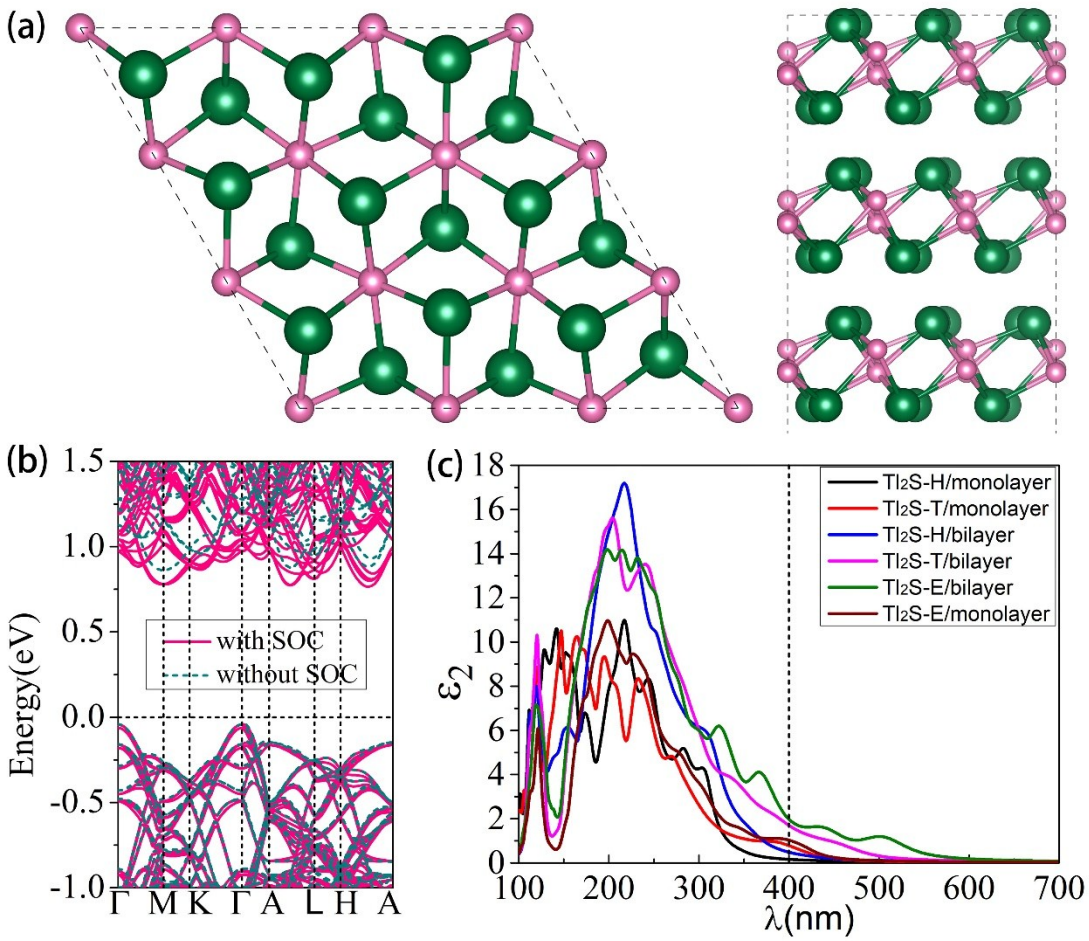

Figure S1. The top (a) and side (b) views of the crystal structures of bulk E-Tl_2S. Band structures of (c) bulk E-Tl_2S calculated using PBE and PBE+SOC. (d) Calculated absorption spectra for ML and BL X-Tl_2S (X=E, 1T, 2H) of the 2D structures at HSE06 level.

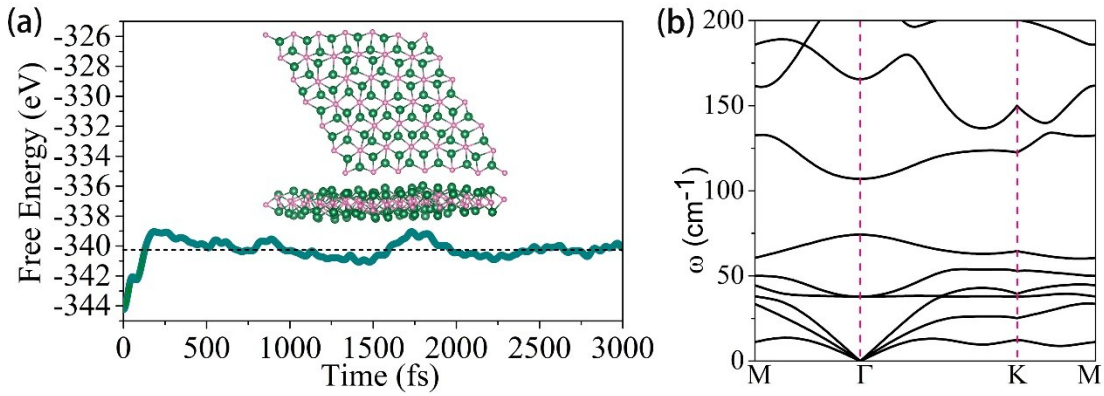

Figure S2. (a) Variation of total energy at 1 ps during ab initio MD simulation at a temperature of 300 K. And the insets show the corresponding snapshots at the end of MD simulation from side and top views. (b) Phonon band dispersion ML 1T-Tl_2S.

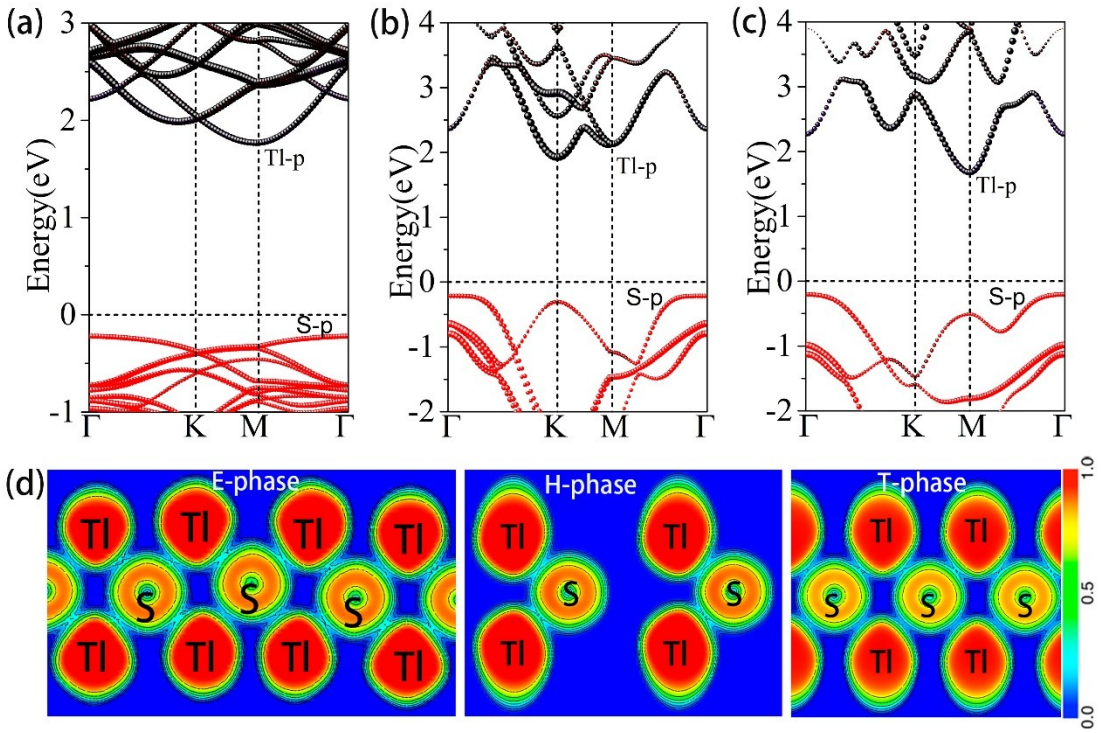

Figure S3. Orbit-projected band structures of (a) E-Tl_2S, (b) 2H-Tl_2S, and (c) 1T-Tl_2S near the Fermi level based on HSE06+SOC. The red (black) dots represents the electrical contribution of the Tl p orbitals (S p orbitals). (d) Isosurfaces of electron localization function of X-Tl_2S, with red and blue marking the accumulated and vanishing electron density, respectively.

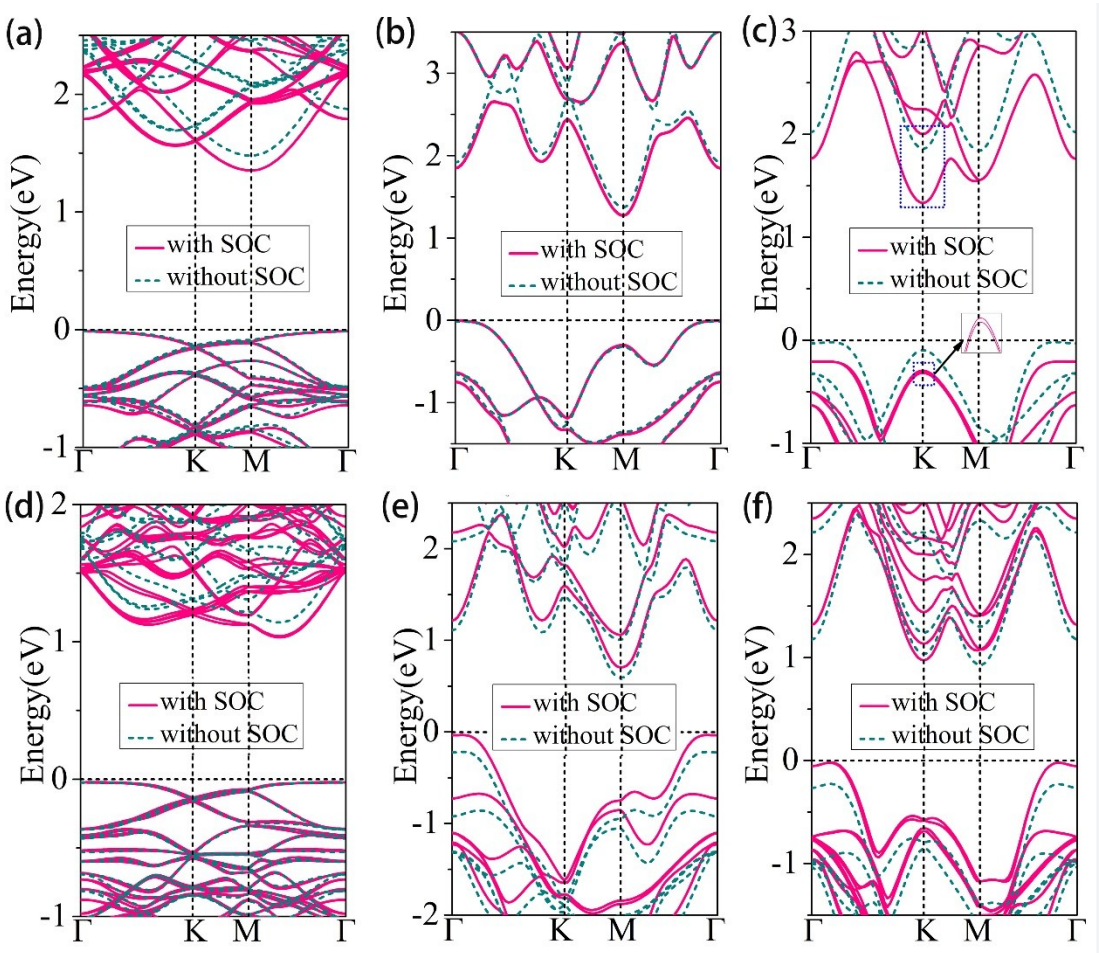

Figure S4. Band structures of (a) ML E-Tl_2S, (b) ML 1T-Tl_2S, (c) ML 2H-Tl_2S, (d) BL E-Tl_2S, (e) BL 1T-Tl_2S, and (f) BL 2H-Tl_2S calculated at PBE and PBE+SOC levels of theory. Solid (red) and dashed (green) curves denote bands with and without SOC, respectively.

Table S1. Effective masses along the x and y Transport Directions in ML 2H-Tl_2S and ML 1T-Tl_2S.

System	HSE06+SOC		PBE+SOC	
	m_{ex}^*	m_{ey}^*	m_{ex}^*	m_{ey}^*
2H-Tl_2S	0.382	0.379	0.405	0.402
1T-Tl_2S	0.265	0.324	0.269	0.327

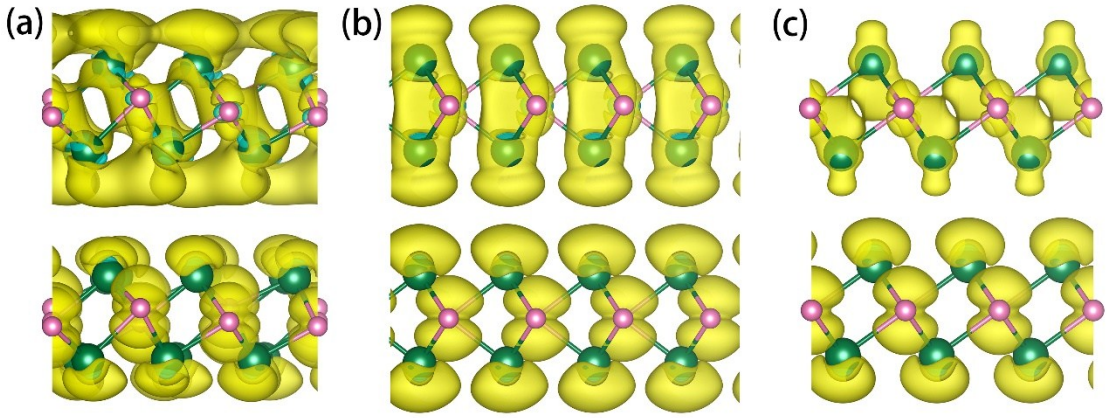

Figure S5. The charge density distribution of the E-Tl_2S (a), 2H-Tl_2S (b), and 1T-Tl_2S (c) at conduction band minimum (above) and valence band maximum (bottom).

HSE results:

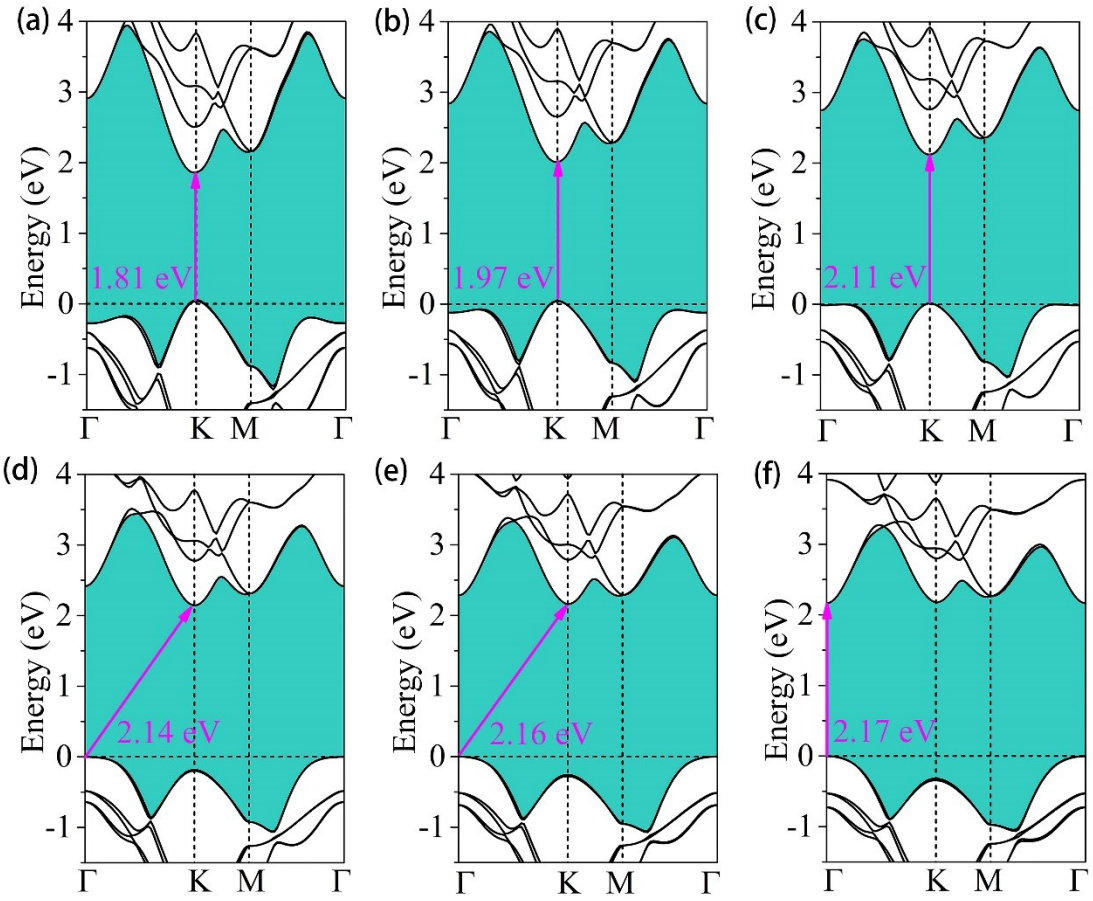

Figure S6. The band structures of 2H-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC.

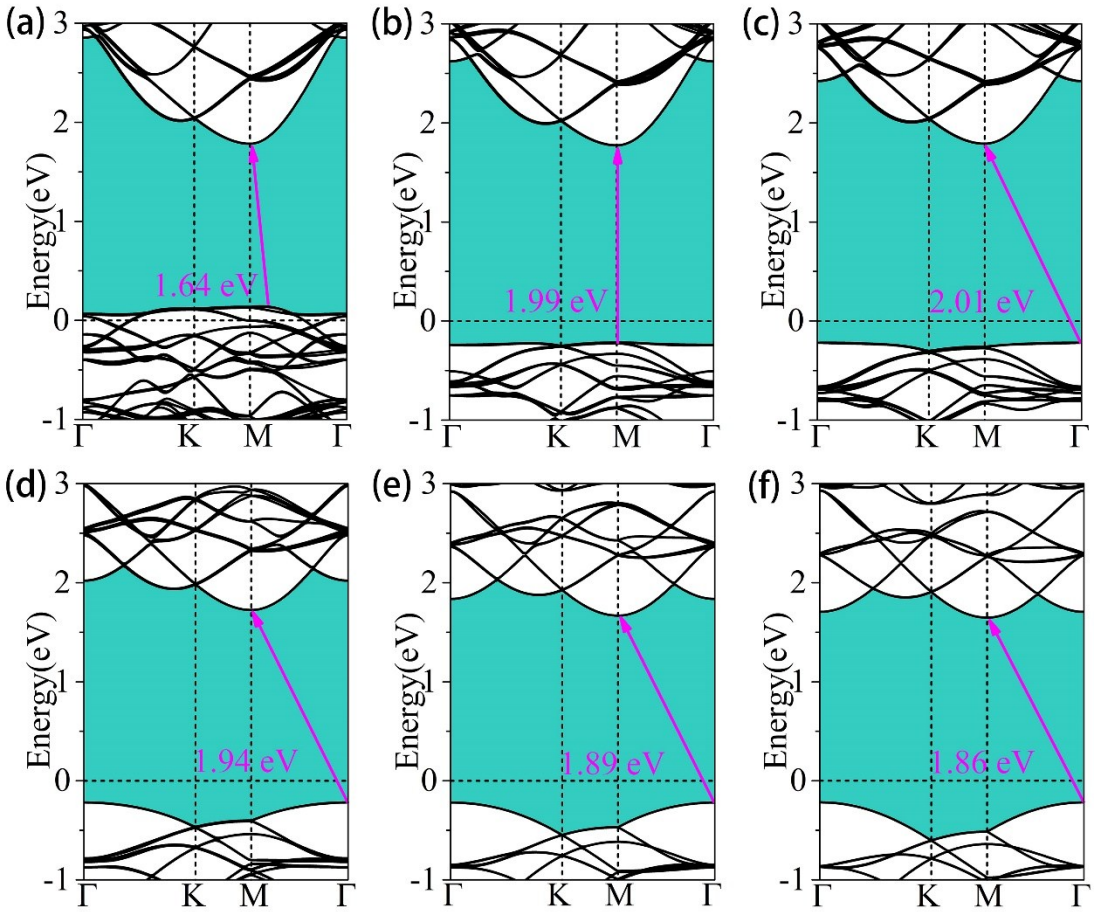

Figure S7. The band structures of E-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC calculated using HSE06+SOC..

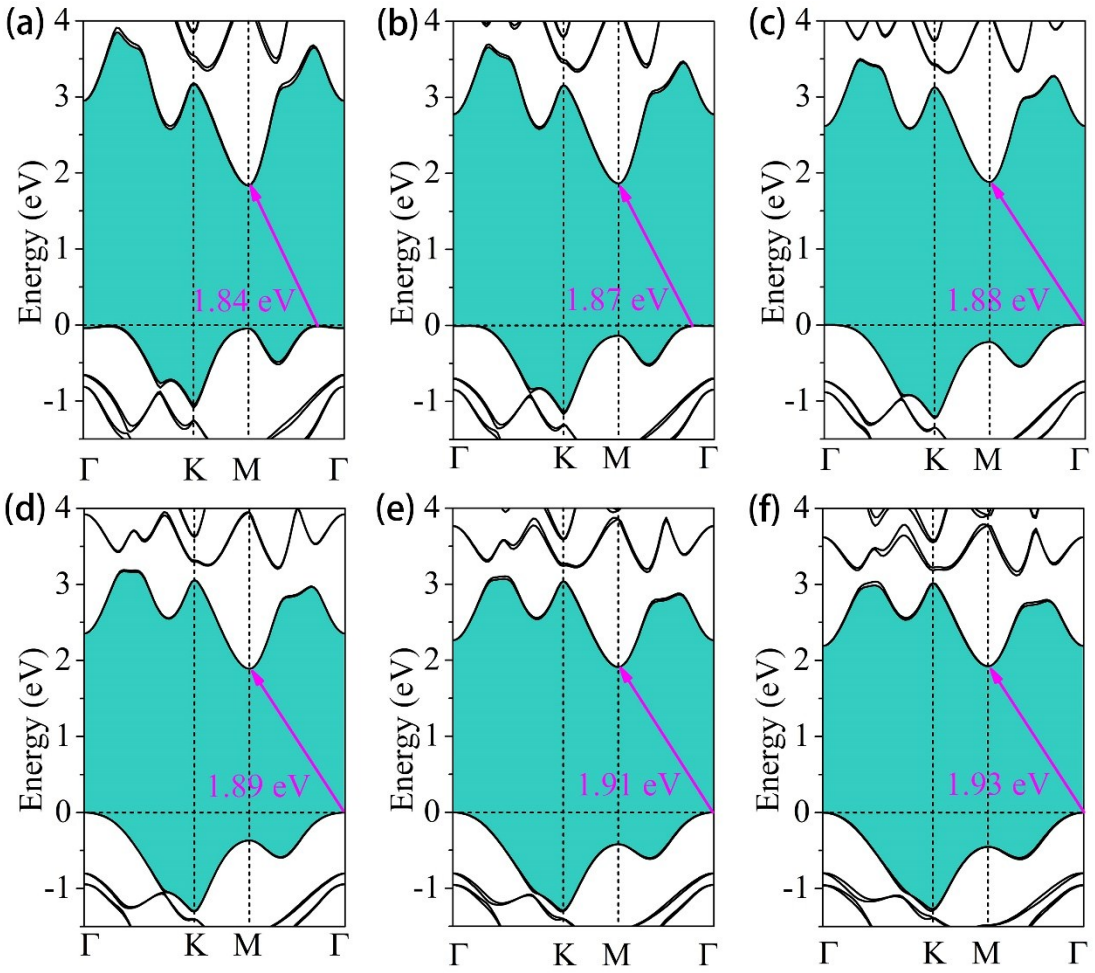

Figure S8. The band structures of 1T-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC calculated using HSE06+SOC..

PBE results:

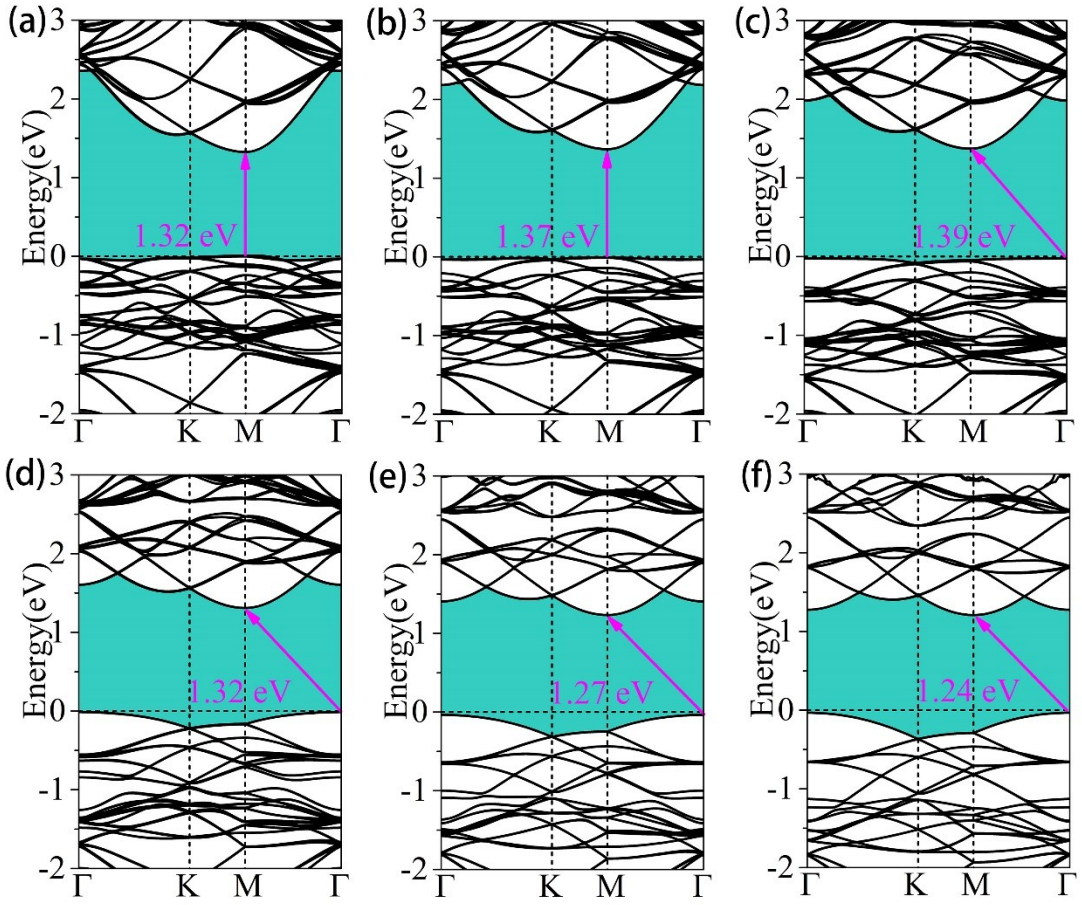

Figure S9. The band structures of E-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC.

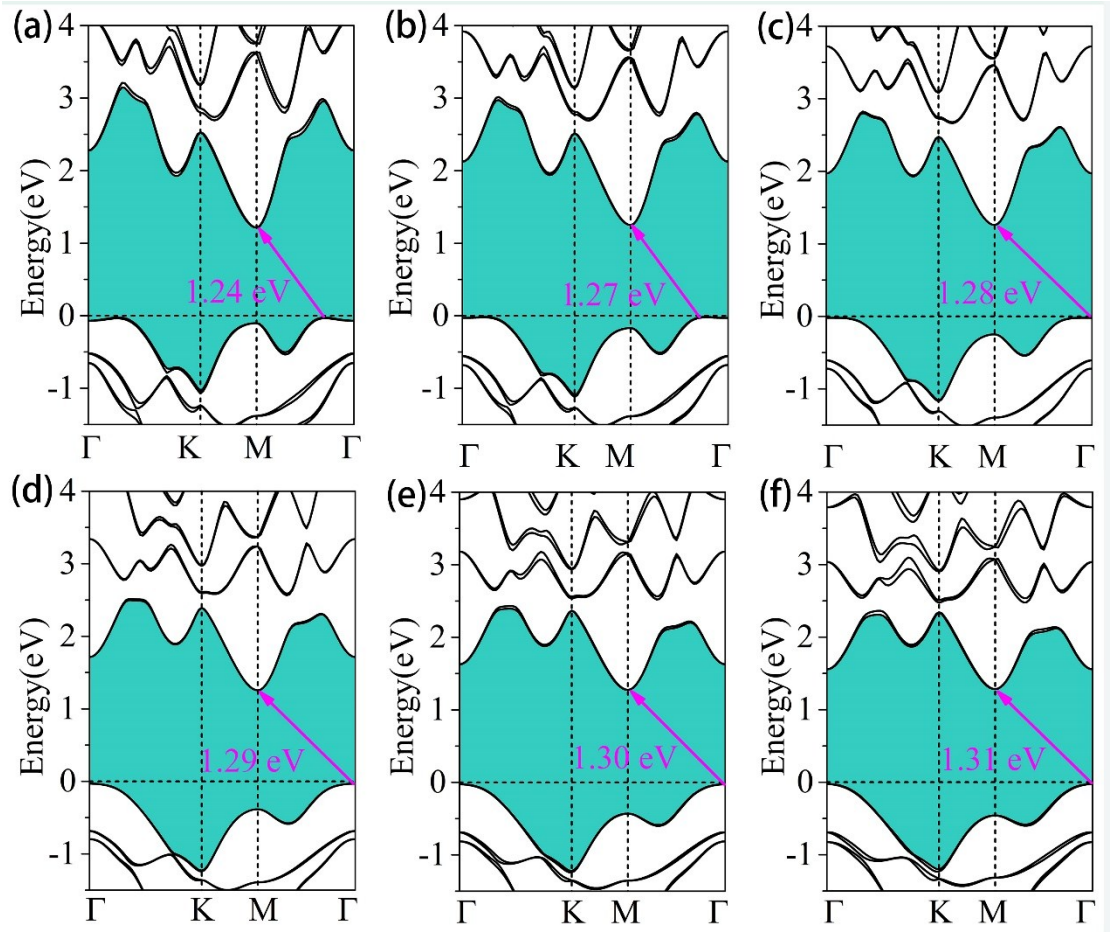

Figure S10. The band structures of 1T-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC.

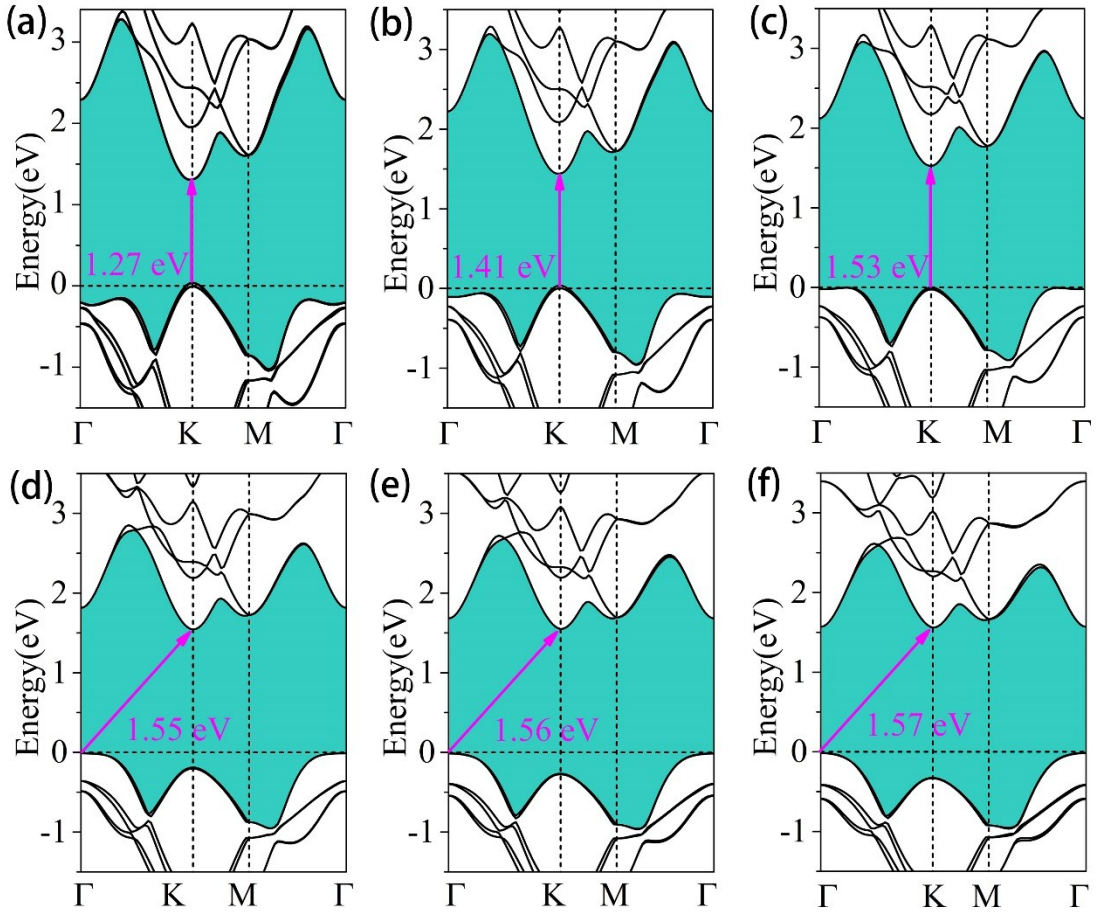

Figure S11. The band structures of 2H-Tl_2S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC.