\( \text{Tl}_2\text{S}: \text{Metal-Shrouded Two-Dimensional Semiconductor} \)

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Figure S1. The top (a) and side (b) views of the crystal structures of bulk E-Tl$_2$S. Band structures of (c) bulk E-Tl$_2$S calculated using PBE and PBE+SOC. (d) Calculated absorption spectra for ML and BL X-Tl$_2$S (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$_2$S.
Figure S3. Orbit-projected band structures of (a) E-Tl₂S, (b) 2H-Tl₂S, and (c) 1T-Tl₂S 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-Tl₂S, with red and blue marking the accumulated and vanishing electron density, respectively.
Figure S4. Band structures of (a) ML E-Tl$_2$S, (b) ML 1T-Tl$_2$S, (c) ML 2H-Tl$_2$S, (d) BL E-Tl$_2$S, (e) BL 1T-Tl$_2$S, and (f) BL 2H-Tl$_2$S 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-Tl₂S and ML 1T-Tl₂S.

<table>
<thead>
<tr>
<th>System</th>
<th>HSE06+SOC</th>
<th>PBE+SOC</th>
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<tbody>
<tr>
<td></td>
<td>$m_{ex}^*$</td>
<td>$m_{ey}^*$</td>
</tr>
<tr>
<td>2H-Tl₂S</td>
<td>0.382</td>
<td>0.379</td>
</tr>
<tr>
<td>1T-Tl₂S</td>
<td>0.265</td>
<td>0.324</td>
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</table>

Figure S5. The charge density distribution of the E-Tl₂S (a), 2H-Tl₂S (b), and 1T-Tl₂S (c) at conduction band minimum (above) and valence band maximum (bottom).
HSE results:

**Figure S6.** The band structures of 2H-Tl$_2$S 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$_2$S 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-Tl₂S 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-TI₂S 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-Tl₂S 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$_2$S under different strain of (a) -3%, (b) -2%, (c) -1%, (d) 1%, (e) 2% and (f) 3% with SOC.