Supporting information for

Self-Powered SnS$_{1-x}$Se$_x$ Alloy/Silicon Heterojunction Photodetectors with High Sensitivity in a Wide Spectral Range

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Method S1| Density Functional Theory (DFT) calculations

The first principles calculations in this work were performed by the VASP code based on DFT. The projected augmented wave (PAW) pseudopotentials with an energy cutoff of 500 eV were employed. Perdew-Burke-Ernzerhofer (PBE) functional with general gradient approximation exchange correlation was used for the structure optimization of SnS, SnSe monolayers and their alloys. A vacuum larger than 12 Å is used to eliminate the interaction between adjacent slabs. In addition, the convergence criteria for total energy is chosen as $10^{-5}$ eV and all the atoms and the lattice vectors were fully relaxed until the force on each atom is less than 0.01 eV · Å$^{-1}$. The SnS$_{1-x}$Se$_x$ alloys are assumed to be random and are constructed by the special quasirandom structures in a supercell containing 36 atoms. All the supercells are optimized with k-point sampling of $4 \times 4 \times 1$. 
Figure S1. Optical images of other as-prepared SnS$_{1-x}$Se$_x$ ($0 \leq x \leq 1$) nanosheets on the SiO$_2$/Si substrates by NGPVD method: (a) (b) (c) 5x magnification. (d) (e) (f) 20x magnification. The scale bar is 100 μm.
Figure S2. AFM images of the as-prepared Sn$_{1-x}$Se$_x$ (0 ≤ x ≤ 1) nanosheets: (a) SnS. (b) SnS$_{0.75}$Se$_{0.25}$. (c) SnS$_{0.25}$Se$_{0.75}$. (d) SnSe.
Figure S3. XRD profiles of the as-prepared SnS$_{1-x}$Se$_x$ (0 ≤ x ≤ 1) nanosheets.
Figure S4. Electrical and optoelectrical properties of the as-prepared SnS photodetector: (a) Transfer curve of the photodetector under dark condition. Inset is the optical image of the device. The scale bar is 5 μm. (b) $I_{ds}$-$V_{ds}$ curve under dark condition. (c) Photocurrent as a function of the incident light density at $V_{ds} = 1$ V. (d) Responsivity and photocurrent as a function of light power density at $V_{ds} = 1$ V. (e) Photo-response curve under 635 nm illumination. ($P = 47.59$ mW·cm$^{-2}$) (f) Rising and decay time of the photodetector.
Figure S5. Electrical and optoelectrical properties of the as-prepared SnS$_{0.75}$Se$_{0.25}$ photodetector: (a) Transfer curve of the photodetector under dark condition. Inset is the optical image of the device. The scale bar is 5 μm. (b) $I_{ds}$-$V_{ds}$ curve under dark condition. (c) Photocurrent as a function of the incident light density at $V_{ds} = 1$ V. (d) Responsivity and photocurrent as a function of light power density at $V_{ds} = 1$ V. (e) Photo-response curve under 635 nm illumination. ($P = 47.59$ mW·cm$^{-2}$) (f) Rising and decay time of the photodetector.
Figure S6. Electrical and optoelectrical properties of the as-prepared SnS$_{0.5}$Se$_{0.5}$ photodetector: (a) Transfer curve of the photodetector under dark condition. Inset is the optical image of the device. The scale bar is 10 μm. (b) $I_{ds}$-$V_{ds}$ curve under dark condition. (c) Photocurrent as a function of the incident light density at $V_{ds} = 1$ V. (d) Responsivity and photocurrent as a function of light power density at $V_{ds} = 1$ V. (e) Photo-response curve under 635 nm illumination. ($P = 47.59$ mW·cm$^{-2}$) (f) Rising and decay time of the photodetector.
Figure S7. Electrical and optoelectrical properties of the as-prepared SnSe photodetector: (a) Transfer curve of the photodetector under dark condition. Inset is the optical image of the device. The scale bar is 5 μm. (b) $I_d$-$V_d$ curve under dark condition. (c) Photocurrent as a function of the incident light density at $V_d$ = 1 V. (d) Responsivity and photocurrent as a function of light power density at $V_d$ = 1 V. (e) Photo-response curve under 635 nm illumination. ($P = 47.59$ mW·cm$^{-2}$) (f) Rising and decay time of the photodetector.
Figure S8. Electrical and optoelectrical properties of the as-prepared SnS$_{0.25}$Se$_{0.75}$ photodetector: (a) $I_{ds}$-$V_{ds}$ curve under dark condition. (b) Photocurrent as a function of the incident light density at $V_{ds} = 1$ V. (c) Photo-response curve under 635 nm illumination. ($P = 44.88$ mW·cm$^{-2}$) (d) Rising and decay time of the photodetector.
Figure S9. Raman intensity mapping of the SnS$_{0.25}$Se$_{0.75}$ nanosheets on SiO$_2$/Si substrate. (a) optical image of the nanosheets, red rectangle is the measured area. The scale bar is 5 μm. (b) Alloy-like $A_g$ (71 cm$^{-1}$). (c) SnSe-like $B_{3g}$ (105 cm$^{-1}$). (d) SnS-like $A_g$ (193 cm$^{-1}$). The measurement was performed with 532 nm laser excitation at room temperature.
Figure S10. HRTEM-EDS of Sn$_{0.25}$Se$_{0.75}$ nanosheets. (a) TEM image at low
magnification. (b) Sn, (c) S, (d) Se element mapping image. (e) The corresponding EDS spectrum of SnS$_{0.25}$Se$_{0.75}$ nanosheets in the yellow square called Area 1. (f) Table of the extracted EDS data from (a).

Figure S11. Comparison of zero-bias operational photo-response curves of the as-prepared SnS$_{0.25}$Se$_{0.75}$/Si photodetector as a function of light power density from 0.03 to 0.25 mW·cm$^{-2}$ under 635 nm illumination.
Figure S12. Zero-bias operational photo-switching on-off curves of the as-prepared SnS<sub>0.25</sub>Se<sub>0.75</sub>/Si photodetector as a function of light power density under 635 nm: (a) 0.03 mW·cm<sup>-2</sup>, (b) 0.06 mW·cm<sup>-2</sup>, (c) 0.12 mW·cm<sup>-2</sup>, (d) 0.25 mW·cm<sup>-2</sup>, (e) 0.74 mW·cm<sup>-2</sup>, (f) 9.60 mW·cm<sup>-2</sup>, (g) 24.27 mW·cm<sup>-2</sup>, (h) 44.88 mW·cm<sup>-2</sup>, (i) 118.71 mW·cm<sup>-2</sup>. 
Figure S13. Photocurrent of the as-prepared SnS$_{0.25}$Se$_{0.75}$/Si photodetector as a function of light power density at $V_{ds} = 0$ V under 635 nm illumination.

Figure S14. Zero-bias operational photo-switching on-off curves of the as-prepared SnS$_{0.25}$Se$_{0.75}$/Si photodetector as a function of light power density.
under 808 nm: (a) 0.12 mW·cm⁻², (b) 0.24 mW·cm⁻², (c) 0.45 mW·cm⁻², (d) 0.89 mW·cm⁻², (e) 1.79 mW·cm⁻², (f) 9.15 mW·cm⁻², (g) 22.52 mW·cm⁻², (h) 39.79 mW·cm⁻², (i) 119.37 mW·cm⁻².

Figure S15. Responsivity and photo-current as a function of light power density under 808 nm illumination.

Figure S16. Zero-bias operational photo-switching on-off curves of the as-prepared SnS₀.₂₅Se₀.₇₅/Si photodetector under different wavelength: (a) 1000 nm. (b) 1100 nm.
Table S1 Comparison of figures-of-merit for self-driven photodetectors based on vdWs heterostructure.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Wavelength (nm)</th>
<th>Responsivity (mA/W)</th>
<th>EQE (%)</th>
<th>D* (Jones)</th>
<th>Response time</th>
<th>( I_{\text{light}}/I_{\text{dark}} )</th>
<th>Reference</th>
</tr>
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<tbody>
<tr>
<td>SnS(<em>{0.25})Se(</em>{0.75})/Si</td>
<td>635</td>
<td>377</td>
<td>73.8</td>
<td>~10(^{11})</td>
<td>4.7/3.</td>
<td>4.5×10(^2)</td>
<td>This work</td>
</tr>
<tr>
<td>SnS/SnS(_2)</td>
<td>405</td>
<td>/</td>
<td>0.13</td>
<td>/</td>
<td>/</td>
<td>/</td>
<td>8</td>
</tr>
<tr>
<td>PtSe(_2)/GaAs</td>
<td>808</td>
<td>262</td>
<td>/</td>
<td>10(^{12})</td>
<td>5.5/6.</td>
<td>10(^4)</td>
<td>9</td>
</tr>
<tr>
<td>MoS(_2)/GaAs</td>
<td>635</td>
<td>321</td>
<td>/</td>
<td>3.5×10(^{13})</td>
<td>17/31</td>
<td>/</td>
<td>10</td>
</tr>
<tr>
<td>MoS(_2)/h-BN/graphene</td>
<td>532</td>
<td>360</td>
<td>80</td>
<td>6.7×10(^{10})</td>
<td>/</td>
<td>10(^3)</td>
<td>11</td>
</tr>
<tr>
<td>CdS/MoTe(_2)</td>
<td>White light</td>
<td>55</td>
<td>15.0</td>
<td>1</td>
<td>50/50</td>
<td>/</td>
<td>12</td>
</tr>
<tr>
<td>SnS/Si</td>
<td>400</td>
<td>17</td>
<td>/</td>
<td>8.1×10(^{10})</td>
<td>24.9</td>
<td>10(^4)</td>
<td>13</td>
</tr>
<tr>
<td>Graphene/Si</td>
<td>850</td>
<td>0.435</td>
<td>/</td>
<td>10(^9)</td>
<td>1.3/3</td>
<td>10(^4)</td>
<td>14</td>
</tr>
<tr>
<td>Se/ZnO</td>
<td>370</td>
<td>2.65</td>
<td>/</td>
<td>/</td>
<td>0.69/1</td>
<td>10(^4)</td>
<td>15</td>
</tr>
<tr>
<td>ITO/SnS/Si/Al</td>
<td>850</td>
<td>8.3</td>
<td>/</td>
<td>5.3×10(^9)</td>
<td>34/15.</td>
<td>21.4</td>
<td>16</td>
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<tr>
<td>MoS(_2)/Si</td>
<td>880</td>
<td>300</td>
<td>/</td>
<td>10(^{13})</td>
<td>3/40μs</td>
<td>/</td>
<td>17</td>
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</table>

Reference


