## Visible-to-near-infrared photodetectors based on SnS/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> p–n heterostructures with a fast response speed and high normalized detectivity

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Fig. S1 (a) Diagram of the photodetector based on SnS/SnSe<sub>2</sub> heterostructure.

Fig. S1 (a) shows the diagram of the photodetector and the size of the device is nearly 550  $\times$  550  $\mu$ m<sup>2</sup>.



Fig. S2. Output curves of photodetectors based on SnSe<sub>2</sub> (a), SnS (b), and SnSe (c). Transfer curves of photodetectors based on SnSe<sub>2</sub> (d), SnS (e), and SnSe (f). Schematic images are shown in the insets.

The electrical characteristics of the photodetectors based on individual materials were investigated. Figs. S2 (a–c) shows the output curves of photodetectors based on SnSe<sub>2</sub>, SnS, and SnSe, respectively. The output curves show good linearity, implying a good ohmic contact between samples and Au electrodes. The transfer curves of photodetectors based on SnSe<sub>2</sub>, SnS, and SnSe are shown in Figs. S2 (d–f). For SnSe<sub>2</sub>, the drain–source current ( $I_{ds}$ ) increases with the increase of  $V_{gs}$ , indicating that SnSe<sub>2</sub> thin films show n–type behavior. Additionally, SnS and SnSe thin films exhibit p–type behaviors. The mobility values of photodetectors based on SnSe<sub>2</sub>, SnS, and SnSe were estimated to be 10, 5.29, and 6.84 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>.



Fig. S3. photodetection performance of photodetectors based individual materials under the illumination of 405 nm. Current–voltage curves of photodetectors based on SnSe<sub>2</sub> (a), SnS (b), and SnSe (c). Transient photoresponse of photodetectors based on SnSe<sub>2</sub> (d), SnS (e), and SnSe (f). Response time of photodetectors based on SnSe<sub>2</sub> (g), SnS (h), and SnSe (i).

The photodetection performance of photodetectors based on SnSe<sub>2</sub>, SnS, and SnSe under the illumination of 405 nm were detected. Figs. S3 (a–c) shows the current–voltage ( $I_{ds}-V_{ds}$ ) curves of photodetectors based on individual materials with different power densities of 17.8, 35.7, 75.3 µW mm<sup>-2</sup>. As the light power density increased,  $I_{ds}$  increased simultaneously. Figs. S2 (d–f) presents the current–time ( $I_{ds}$ –T) curves of such photodetectors at  $V_{ds} = 5$  V. As photodetectors were exposed to illumination, the current increased to saturation. While the laser was turned off, the current decayed immediately to the initial state. The response time of such photodetectors was shown in Figs. S3 (g–i). The response time including the rise time  $\tau_r$  and fall time  $\tau_f$  were estimated as 17.97/19.43 ms (SnSe<sub>2</sub>), 31.96/34.89 ms (SnS), and 53.65/60.88 ms (SnSe).



Fig. S4. Photocurrent as a function of optical power density under the illumination of 405 (a) and 850 nm (b).

Fig. S4 depicts the relationship between photocurrent and optical power density. Under the 405 nm illumination, the  $\alpha$  values were estimated to be 0.69 and 0.60 for the SnSe<sub>2</sub>/SnS and SnSe<sub>2</sub>/SnSe photodetector, which are larger than that of the photodetector based on the SnSe<sub>2</sub> thin layers (0.32). Similarly, under the 850 nm illumination, the  $\alpha$  values of SnSe<sub>2</sub>/SnS (0.97) and SnSe<sub>2</sub>/SnSe (0.95) photodetector are also larger than that of SnSe<sub>2</sub>—based photodetectors (0.77), revealing the enhanced photocurrent conversion efficiency.



Fig. S5. photodetection performance of SnS/SnSe<sub>2</sub> photodetectors under the illumination of different wavelengths. Current–voltage curves of SnS/SnSe<sub>2</sub> photodetectors to 532 (a), 650 (b), and 850 nm (c). Transient photoresponse of SnS/SnSe<sub>2</sub> photodetectors to 532 (d), 650 (e), and 850 nm (f).

The photodetection performance of SnS/SnSe<sub>2</sub> photodetectors under the illumination of different wavelengths were detected. Figs. S5 (a–c) shows the current–voltage ( $I_{ds}-V_{ds}$ ) curves of SnS/SnSe<sub>2</sub> photodetectors under the illumination of 532, 650, and 850 nm. The photodetectors exhibited an excellent broad–spectrum photodetection performance in the region from 532 nm to 850 nm. Figs. S5 (d–f) presents the current–time ( $I_{ds}-T$ ) curves of SnS/SnSe<sub>2</sub> photodetectors with different wavelength. The bias voltage was set at 5 V. As can be seen that currents could alternate between saturation and initial state rapidly. And photocurrents increased multiplicatively as the light power density increased.



Fig. S6. photodetection performance of SnSe/SnSe<sub>2</sub> photodetectors under the illumination of different wavelengths.  $I_{ds}-V_{ds}$  curves of SnSe/SnSe<sub>2</sub> photodetectors to 532 (a), 650 (b), and 850 nm (c).  $I_{ds}-T$  curves of SnSe/SnSe<sub>2</sub> photodetectors to 532 (d), 650 (e), and 850 nm (f).

The photodetection performance of SnSe/SnSe<sub>2</sub> photodetectors under the illumination of different wavelengths were detected. Figs. S6 (a–c) presents the  $I_{ds}-V_{ds}$  curves of SnSe/SnSe<sub>2</sub> photodetectors under the illumination of 532, 650, and 850 nm. The SnSe/SnSe<sub>2</sub> photodetectors also showed an excellent photodetection performance in the region from 532 nm to 850 nm. Figs. S6 (d–f) plots the  $I_{ds}-T$  curves of SnS/SnSe<sub>2</sub> photodetectors with different wavelength. The photodetectors based on SnS/SnSe<sub>2</sub> also exhibit an instant response to the broad–spectrum illumination.



As shown in Figs. S7 (a) and (b), the noise currents of the  $SnS/SnSe_2$  and  $SnSe/SnSe_2$  photodetectors can be derived from the fast Fourier transform of the dark current<sup>[1]</sup>. In general, the photodetector noise consists of the flicker noise (1/f noise), thermal noise, and shot noise<sup>[2]</sup>. At the frequency below 14.7 Hz (12.5 Hz), the noise current is mainly flicker noise and has a linear relationship with the frequency. At the high frequency above 14.7 Hz (12.5 Hz), the noise current is dominated by the thermal noise and shot noise dominate.



Fig. S8. Responsivity (a) and normalized detectivity (b) as a function of the power intensity of the 405 nm illumination.

Figs. S8 (a) and (b) show the responsivity (*R*) and normalized detectivity ( $D^*$ ) as a function of the power intensity of the 405 nm illumination. The responsivity and normalized detectivity decrease with the increase of the light power intensity. The maximum *R* values of the SnS/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> photodetectors were determined to be 4.99 × 10<sup>3</sup> and 5.91 × 10<sup>3</sup> A W<sup>-1</sup>, respectively, which are significantly higher than that of photodetectors based on individual SnSe<sub>2</sub> (2.46 × 10<sup>3</sup> A W<sup>-1</sup>), SnS (1.19 × 10<sup>3</sup> A W<sup>-1</sup>), and SnSe (1.37 × 10<sup>3</sup> A W<sup>-1</sup>). The *D*<sup>\*</sup> values of the SnS/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> photodetectors were determined to be 5.78 × 10<sup>12</sup> and 7.03 × 10<sup>12</sup> cm Hz<sup>1/2</sup> W<sup>-1</sup>, which are one order of magnitude higher than that of photodetectors based on individual SnSe<sub>2</sub> (8.79 × 10<sup>11</sup> cm Hz<sup>1/2</sup> W<sup>-1</sup>), SnS (5.69 × 10<sup>11</sup> cm Hz<sup>1/2</sup> W<sup>-1</sup>), and SnSe (9.82 × 10<sup>11</sup> cm Hz<sup>1/2</sup> W<sup>-1</sup>).



Fig. S9. External quantum efficiency (a) and noise equivalent power (b) as a function of the wavelength.

Fig. S9 (a) shows the external quantum efficiency (*EQE*) as a function of the wavelength. The external quantum efficiency decreased with the increase of the wavelength. The maximum *EQE* values of the SnS/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> photodetectors were determined to be  $1.53 \times 10^{4\%}$  and  $1.81 \times 10^{4\%}$ , respectively. Fig. S9 (b) presents the noise equivalent power (*NEP*) as a function of the wavelength. The noise equivalent power increased with the increase of the wavelength. The minimum *NEP* values of the SnS/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> and SnSe/SnSe<sub>2</sub> photodetectors were determined to be  $8.02 \times 10^{-5}$  and  $7.29 \times 10^{-5}$  pW Hz<sup>1/2</sup>, respectively.



Fig. S10. Band structures of the SnSe<sub>2</sub> (a), SnS (b), and SnSe (c).

Furthermore, first–principles calculations based on the density function theory (DFT) were performed using the Vienna Ab–initio Simulation Package (VASP)<sup>[3]</sup>. The generalized gradient approximation (GGA) in the form of Perdew–Burke–Ernzerhof (PBE) was used to illustrate the exchanged–related effects. A vacuum space of 15 Å were used to avoid artificial interactions. The cut–off energy was set to 400 eV for all calculations. The Grimme DFT-D3 scheme was adopted to account for the van der Waals (vdW) interactions <sup>[4][5]</sup>. Energy and force convergence criteria was set to  $10^{-5}$  eV and  $10^{-3}$  eV/Å for the structural optimization. The work function difference between SnSe<sub>2</sub> and SnS is 1.67 eV, which is less than that between SnSe<sub>2</sub> and SnSe (1.73 eV). As shown in Figs. 10 (a–c), the bandgap of SnSe<sub>2</sub>, SnS, and SnSe were calculated to be 0.88, 1.39, and 1.07 eV, respectively.

	$\lambda(nm)$	R (A W <sup>-1</sup> )	$D^*$ (cm Hz <sup>1/2</sup> W)	$\tau_r / \tau_f(\mathrm{ms})$	Reference
SnSe <sub>2</sub>	405	2460.3	8.79×10 <sup>11</sup>	17.97	This work
SnS	405	1390.0	5.69×10 <sup>11</sup>	31.96	This work
SnSe	405	1371.8	9.82×10 <sup>11</sup>	53.65	This work
SnS/SnSe <sub>2</sub>	405	4990.3	$5.80 \times 10^{12}$	3.13	This work
	532	4230.2	$4.84 \times 10^{12}$	4.98	
	650	1750.1	$1.73 \times 10^{12}$	7.22	
	850	939.3	$1.02 \times 10^{12}$	8.76	
SnSe/SnSe <sub>2</sub>	405	5912.3	$7.03 \times 10^{12}$	4.74	This work
	532	5451.8	$5.81 \times 10^{12}$	6.12	
	650	1890.2	$1.63 \times 10^{12}$	8.49	
	850	1102.4	$8.38 \times 10^{11}$	10.37	
$WSe_2/MoS_2$	532	2700	$5.00 \times 10^{11}$	17	ref.[6]
SnTe/Ge	808	0.62	$2.33 \times 10^{10}$	206	ref.[7]
$SnSe_2/MoS_2$	500	9100	$9.3 \times 10^{10}$	200	ref.[8]
$SnSe_2/WSe_2$	532	588	$4.4 \times 10^{10}$	16	ref.[9]
PbSe/MoS <sub>2</sub>	808	19.7	$2.65 \times 10^{10}$	720	ref.[10]
Gr/SnSe <sub>2</sub> /Gr	532	1300	$1.48 \times 10^{12}$	30.2	ref.[11]
PbSe QDs/ZnO	808	0.97	1.86×10 <sup>11</sup>	340	ref.[12]
ZnIn <sub>2</sub> S <sub>4</sub> /Si	808	0.50	$2.00 \times 10^{12}$	29	ref.[13]
Bi <sub>2</sub> Te <sub>3</sub> /Si	635	1	2.50×10 <sup>11</sup>	100	ref.[14]
MoS <sub>2</sub> /SiNWs	450	2.98	$2.70 \times 10^{11}$	450	ref.[15]
$MoS_{2/}CuPc$	500	3000	$2 \times 10^{10}$	0.44	ref.[16]

Tab. S1. A comparison of the photodetecting performance

## **Reference:**

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