

**Supplementary Information**

for

**PtSe<sub>2</sub> Thickness Engineering Towards Fast Response, Large Linear Dynamic Range, and Broadband PtSe<sub>2</sub>/Si Heterojunction Photodetector**

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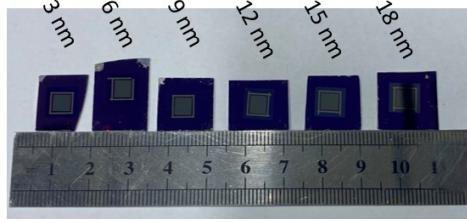
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**Figure S1.** Digital optical images of the devices with various PtSe<sub>2</sub> thicknesses.

### Determination of the diodes' parameters

The diodes' parameters can be obtained from Cheung's functions <sup>1</sup>:

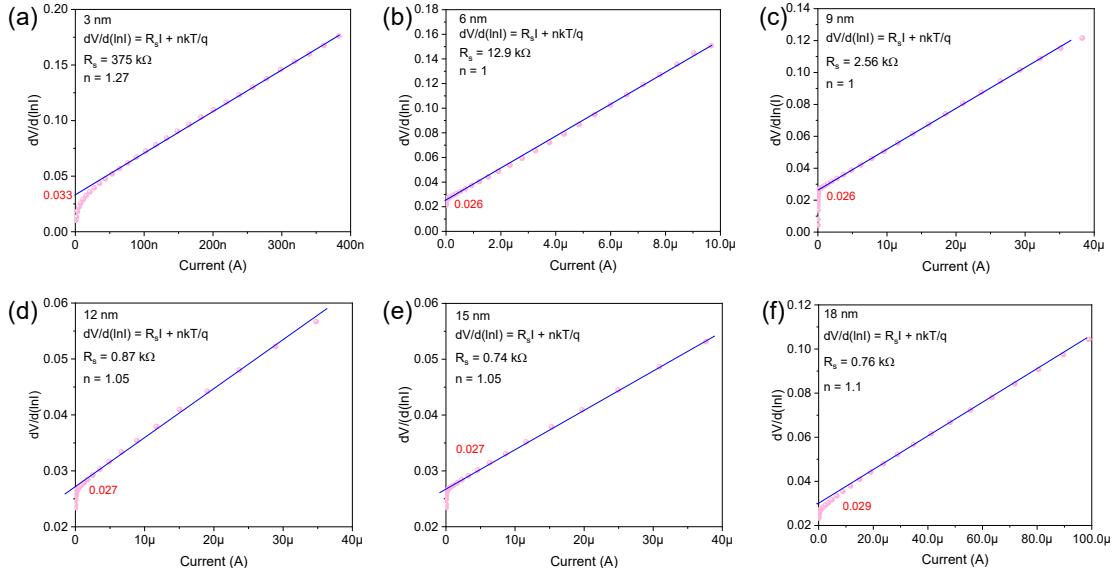
$$\frac{dV}{d\ln I} = IR_s + \frac{n k T}{q} \quad (1)$$

$$H(I) = V - \frac{n k T}{q} \ln \left( \frac{I}{A^* S T^2} \right) \quad (2)$$

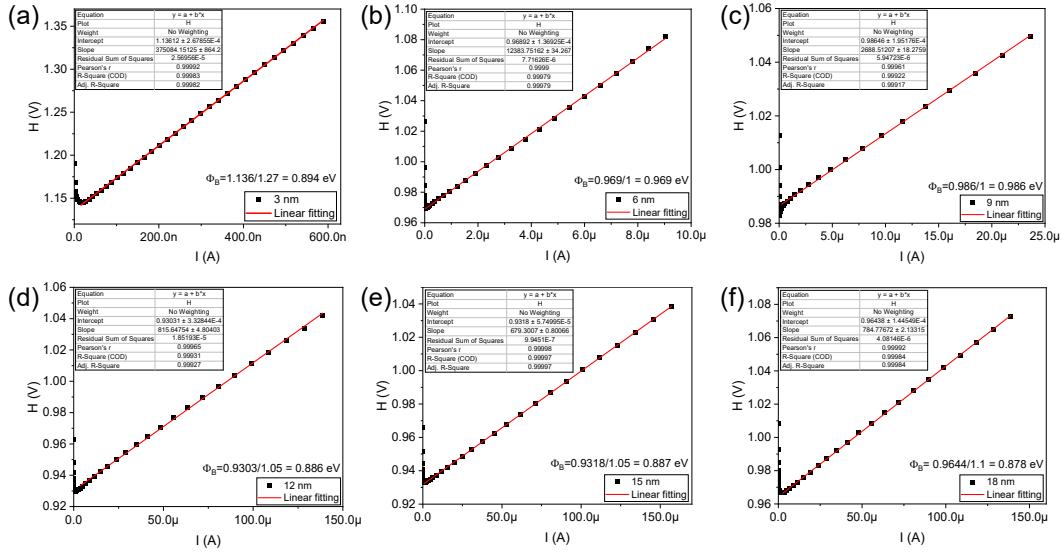
$$H(I) = IR_s + n\Phi_B \quad (3)$$

where  $R_s$  is the series resistance,  $n$  is the ideality factor,  $T$  is the absolute temperature,  $q$  is the elementary charge value,  $A^*$  is the Richardson constant,  $\Phi_B$  is the Schottky barrier height. From the  $dV/d\ln I - I$  plots and linear fittings (**Figure S2**), the  $R_s$  and  $n$  can be evaluated.

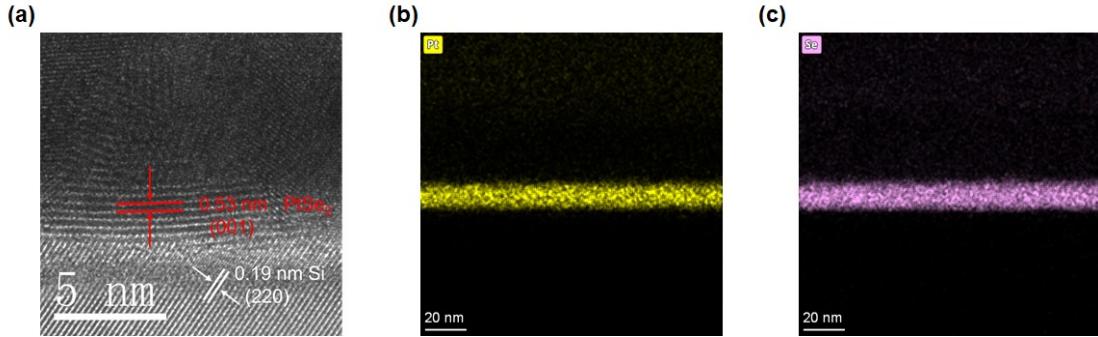
Utilizing the obtained  $n$  to calculate  $H(I)$  according to equation (2), and drawing  $H$  versus  $I$  plots (**Figure S3**), the Schottky barrier heights can be evaluated.



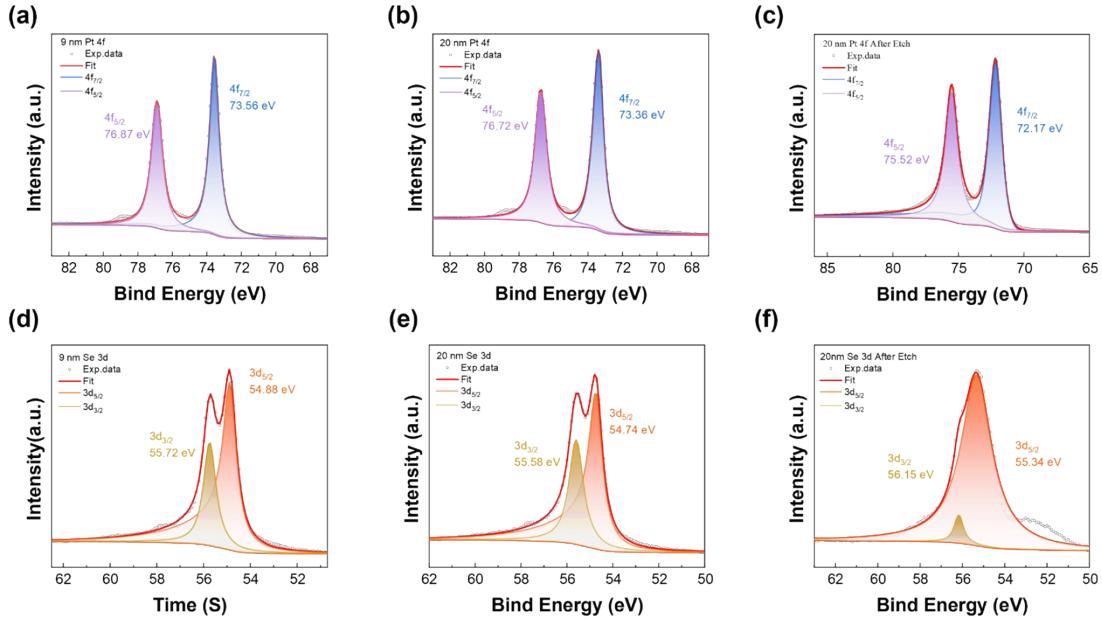
**Figure S2.**  $dV/d\ln I$  versus  $I$  plots to determine  $n$  and  $R_s$ . (a) 3 nm, (b) 6 nm, (c) 9 nm, (d) 12 nm, (e) 15 nm, (f) 18 nm.



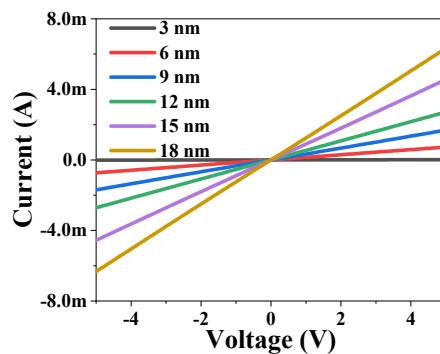
**Figure S3.**  $H$  versus  $I$  plot to determine  $\Phi_B$ . (a) 3 nm, (b) 6 nm, (c) 9 nm, (d) 12 nm, (e) 15 nm, (f) 18 nm.



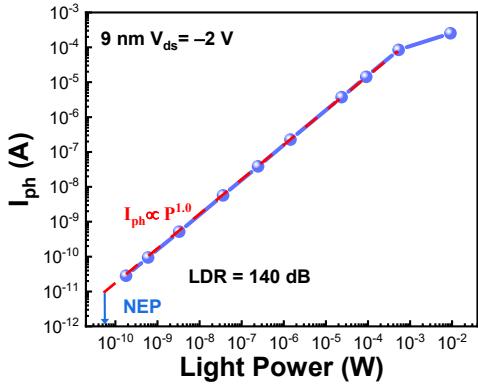
**Figure S4.** HRTEM and EDS mapping. (a) HRTEM image of the heterojunction with a 9 nm-thick  $\text{PtSe}_2$ . EDS mapping: (b) Pt, (c) Se.



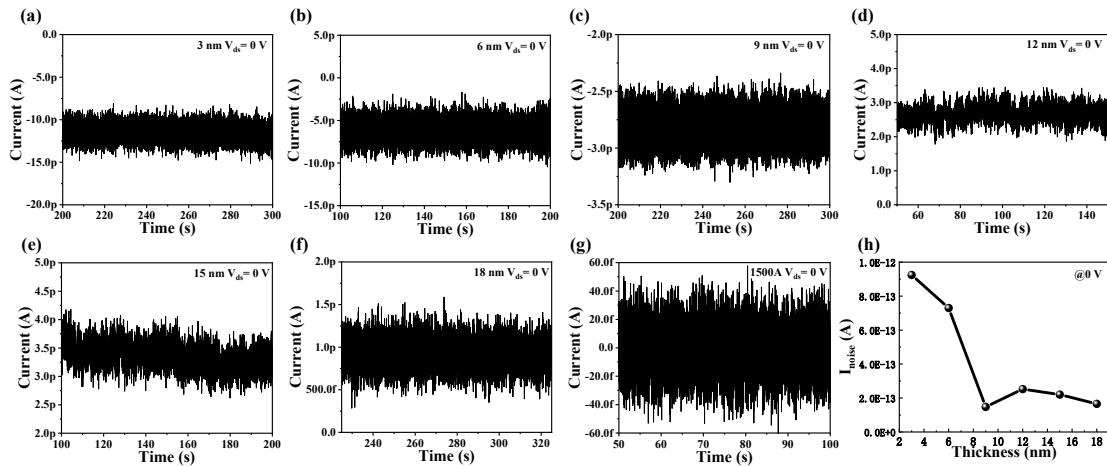
**Figure S5.** The XPS spectra of PtSe<sub>2</sub> with varying thicknesses. The Pt 4f core level: (a) 9 nm PtSe<sub>2</sub>, (b) 20 nm PtSe<sub>2</sub>, (c) 20 nm PtSe<sub>2</sub> after etching 12 nm. The Se 3d core level: (d) 9 nm PtSe<sub>2</sub>, (e) 20 nm PtSe<sub>2</sub>, (f) 20 nm PtSe<sub>2</sub> after etching 12 nm. The 9 nm-thick PtSe<sub>2</sub> film was fully selenized. The surface of the 20 nm-thick PtSe<sub>2</sub> was fully selenized, but within the interior (i.e. 12 nm depth), incomplete selenization was evident. This is indicated by the shifted peaks of the Pt core level and the changes in the profile of the Se core level.



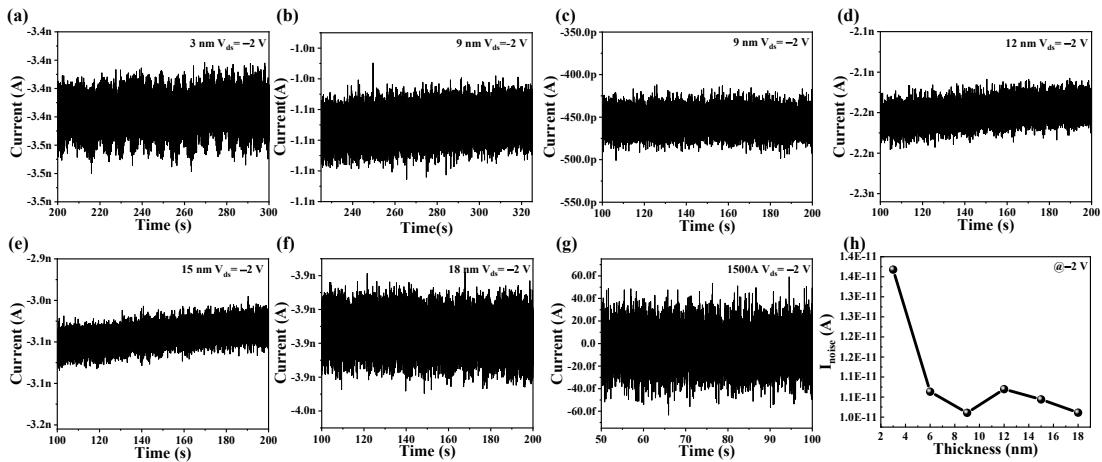
**Figure S6.** IV curves of the PtSe<sub>2</sub> films with various thicknesses.



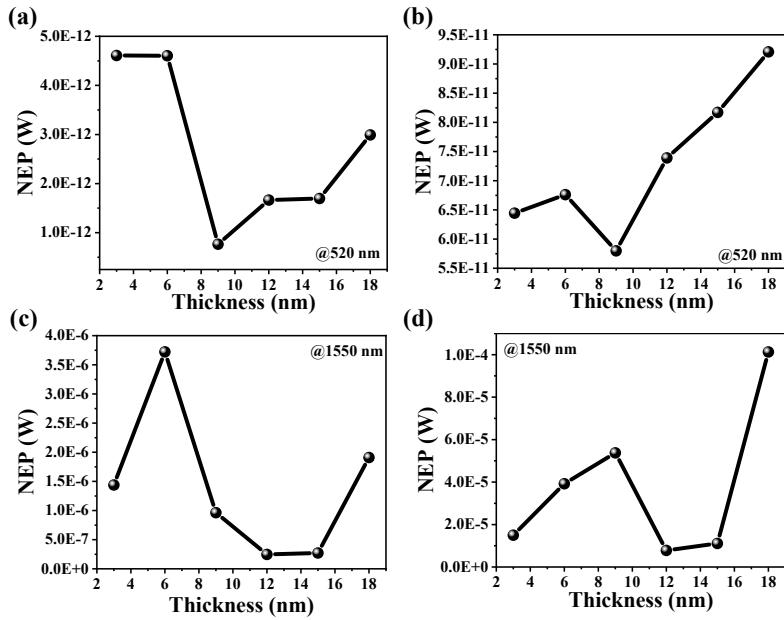
**Figure S7.** Photocurrent versus light power at 520 nm under  $-2$  V bias.



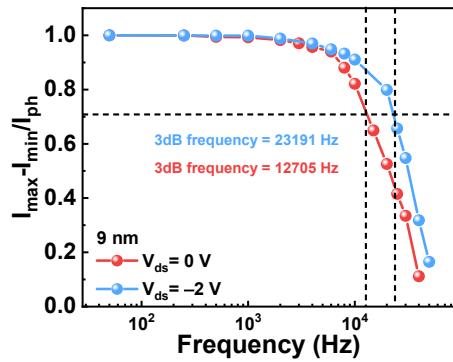
**Figure S8.** Dark-state current as a function of time under 0 V bias. (a) 3 nm, (b) 6 nm, (c) 9 nm, (d) 12 nm, (e) 15 nm, (f) 18 nm, (g) without device, (h) root-mean-square noise current as a function of PtSe<sub>2</sub> film thickness. The none zero average currents are originated from the zero drift or offset from the source-and-measurement unit.



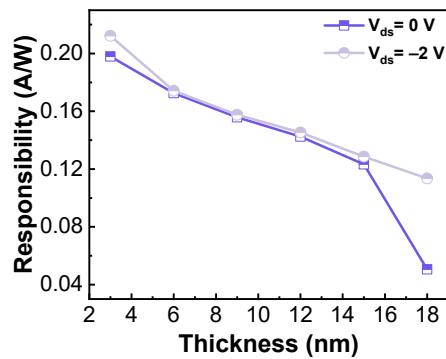
**Figure S9.** Dark-state current as a function of time under  $-2$  V bias. (a) 3 nm, (b) 6 nm, (c) 9 nm, (d) 12 nm, (e) 15 nm, (f) 18 nm, (g) without device, (h) root-mean-square noise current as a function of PtSe<sub>2</sub> film thickness.



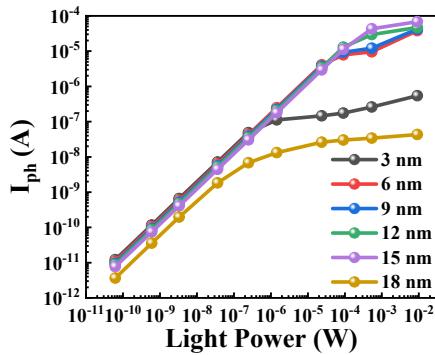
**Figure S10.** NEP as a function of PtSe<sub>2</sub> film thickness. 520 nm: (a) 0 V, (b) -2 V. 1550 nm: (c) 0 V, (d) -2 V.



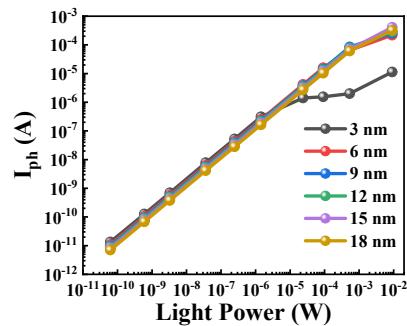
**Figure S11.** 3dB bandwidths at 520 nm under 0 V and -2 V biases.



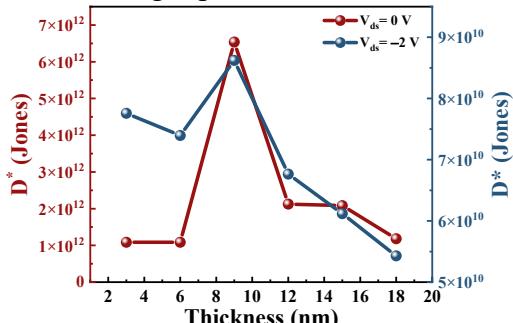
**Figure S12.** Responsivities versus PtSe<sub>2</sub> film thickness at 520 nm under 0 V and -2 V biases.



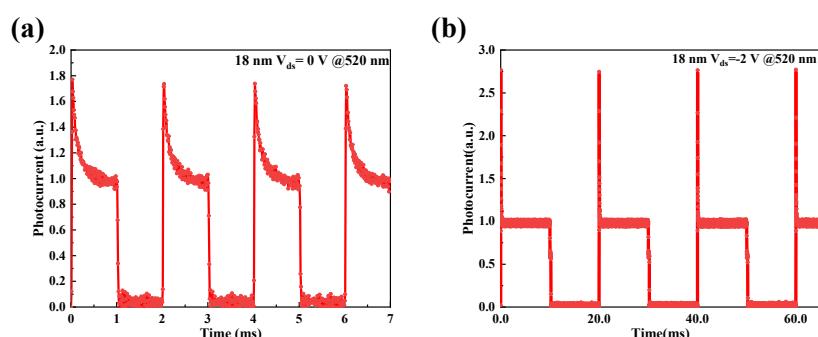
**Figure S13.** photocurrent versus light power at 520 nm under 0 V bias.



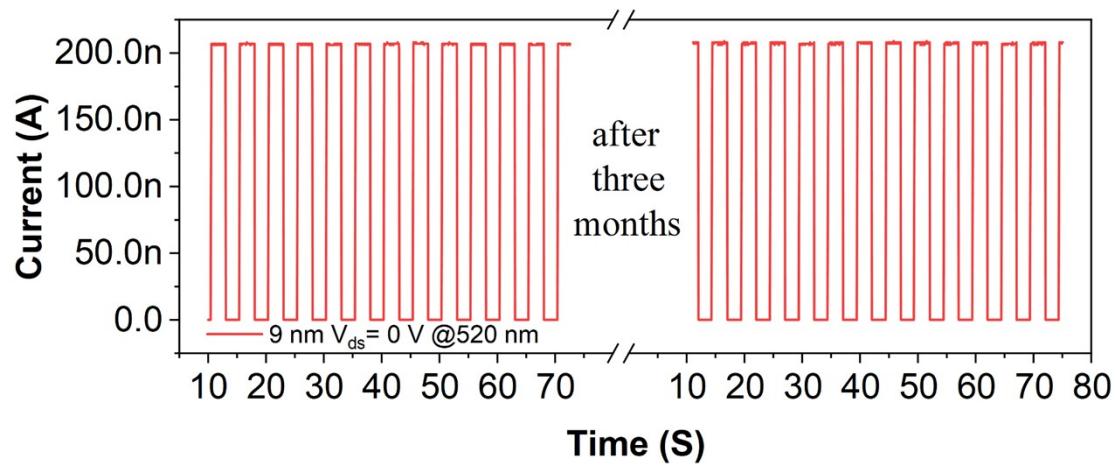
**Figure S14.** Photocurrent versus light power at 520 nm under -2 V bias.



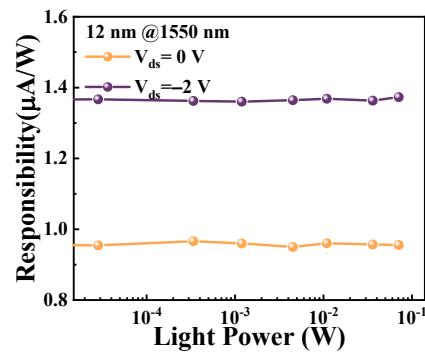
**Figure S15.** Specific detectivity ( $D^*$ ) of the devices with varying  $\text{PtSe}_2$  thickness at 520 nm under 0 V and -2 V biases.



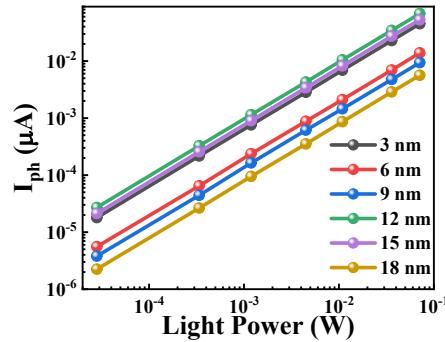
**Figure S16.** Current-time of the device with an 18 nm thick  $\text{PtSe}_2$  layer under rectangular pulsed light illumination. (a) 0 V (b) -2 V.



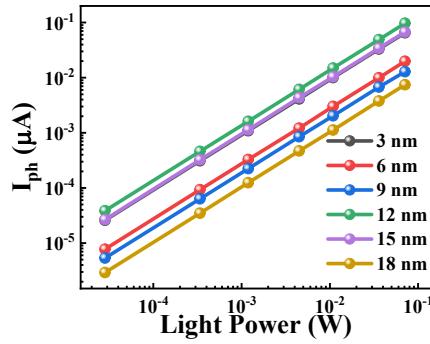
**Figure S17.** Temporal response of the photodetector under the light illumination with alternating on and off cycles.



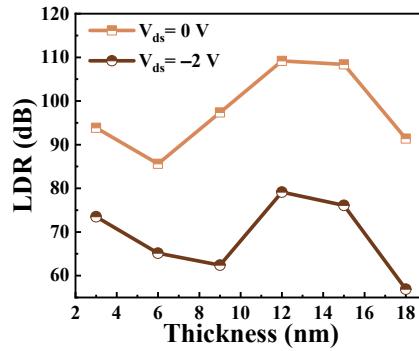
**Figure S18.** Responsivities as a function of light power for device with 12 nm thick  $\text{PtSe}_2$  under 0 V and  $-2$  V biases.



**Figure S19.** Photocurrent versus light power at 1550 nm under 0 V bias.



**Figure S20.** Photocurrent versus light power under  $-2$  V bias.



**Figure S21.** LDRs as a function of PtSe<sub>2</sub> layer thickness at 1550 nm under 0 V and  $-2$  V biases.

**Table S1.** Photodetection performance comparison

Devices	Wavelength h (nm)	Bias (V)	R (A/W)	LDR (dB)	D* (Jones)	Response time ( $\mu$ s)	Ref.
PtSe <sub>2</sub> (9 nm)-Si	520	0	0.16	157	$6.5 \times 10^{12}$	78	Our work
		-2	0.16	139	$8.6 \times 10^{10}$	38	
	800	0	0.24	-	$9.1 \times 10^{12}$	-	
	950	0	0.44	-	$2.4 \times 10^{12}$	-	
PtSe <sub>2</sub> -Si pin	532	-2	0.054	58	$1.35 \times 10^{11}$	2.2/11.8	2
PtSe <sub>2</sub> -Si	808	0	0.52	114	$3.26 \times 10^{13}$	55/170	3
PtSe <sub>2</sub> -Si nanowire array	780	-5	12.65	92	$2.5 \times 10^{13}$	10.1/19.5	4
PtSe <sub>2</sub> /Bi <sub>2</sub> Te <sub>3</sub> /pyramid Si	980	0	0.62	100	$1.37 \times 10^{12}$	0.45/18	5
Graphene/PtSe2/Pyramid Si	980	0	0.528	~80	$4.36 \times 10^{12}$	8.5/10.2	6
PtSe2/ultrathin SiO <sub>2</sub> /Si	808	0	8	158	$4.78 \times 10^{13}$	14.1/15.4-	7
PtSe <sub>2</sub> /graphene/Si	808	-1	0.81	-	$1.29 \times 10^9$	43.6/51.2	8
PtSe <sub>2</sub> /i-Si/n-Si	532	0	0.0465	-	$1.94 \times 10^{11}$	-	9
		-2	0.05	78	$4.78 \times 10^9$	70/290	

**Table S2.** Performance parameters of the PtSe<sub>2</sub>-Si heterojunction photodetectors at 532 nm.

PtSe <sub>2</sub> thickness (nm)	Bias (V)	R (A/W)	D* (Jones)	LDR (dB)	Response times ( $\mu$ s)
3	0	0.198	$1.08 \times 10^{12}$	95	92
	-2	0.212	$7.76 \times 10^{10}$	123	62
6	0	0.172	$1.09 \times 10^{12}$	134	84
	-2	0.174	$7.39 \times 10^{10}$	122	45
9	0	0.155	$6.53 \times 10^{12}$	157	78
	-2	0.157	$8.62 \times 10^{10}$	139	38
12	0	0.142	$2.12 \times 10^{12}$	155	86
	-2	0.145	$6.76 \times 10^{10}$	137	43
15	0	0.123	$2.08 \times 10^{12}$	155	1110
	-2	0.128	$6.12 \times 10^{10}$	136	109
18	0	0.051	$1.18 \times 10^{12}$	61	-
	-2	0.113	$5.43 \times 10^{10}$	135	-

**Table S3.** Performance parameters of the PtSe<sub>2</sub>-Si heterojunction photodetectors at 1550 nm.

PtSe <sub>2</sub> thickness (nm)	Bias (V)	R ( $\mu$ A/W)	D* (Jones)	LDR (dB)	Response times (ms)
3	0	0.63	$3.46 \times 10^6$	95	5.56
	-2	0.91	$3.34 \times 10^5$	123	1.31
6	0	0.20	$1.34 \times 10^6$	134	3.79
	-2	0.27	$1.28 \times 10^5$	123	0.32
9	0	0.14	$5.20 \times 10^6$	157	0.46
	-2	0.19	$9.29 \times 10^4$	139	0.41
12	0	0.95	$1.43 \times 10^7$	155	0.52
	-2	1.36	$6.38 \times 10^5$	137	0.47
15	0	0.76	$1.31 \times 10^7$	155	1.96
	-2	0.96	$4.50 \times 10^5$	136	1.84
18	0	0.08	$1.85 \times 10^6$	61	-
	-2	0.10	$4.94 \times 10^4$	135	-

## References

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