Electronic Supplementary Material (ESI) for Nanoscale.

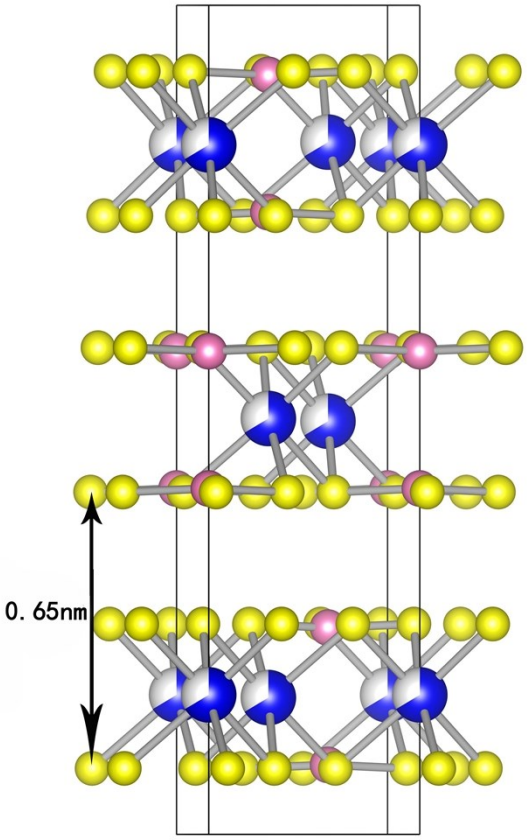
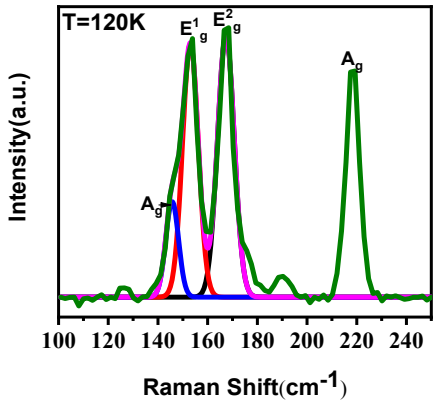


Fig. S1 The side view image of In $In_{4/3}P_2Se_6$



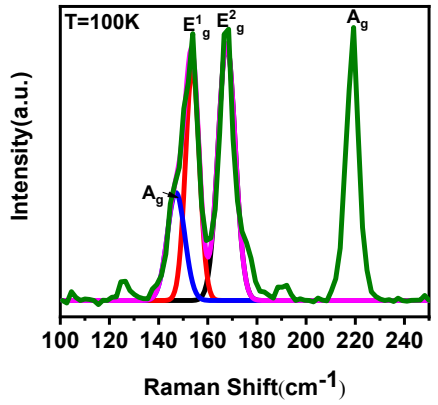


Fig.S2 Raman peaks fitting of $In_{4/3}P_2Se_6$ flakes under 120 and 100 K

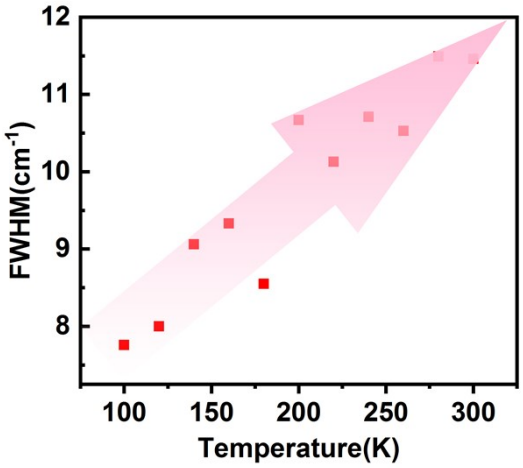


Fig. S3 FWHM of E^2_g as a function of temperature

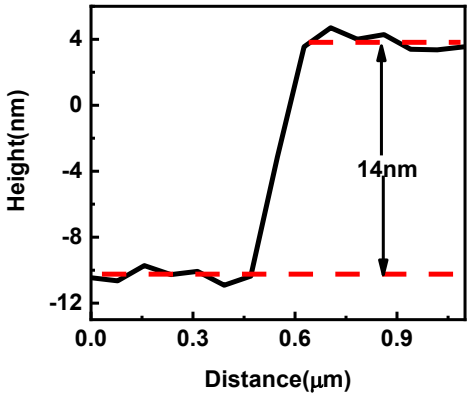


Fig. S4 AFM Height curve of the device in Fig.5b.

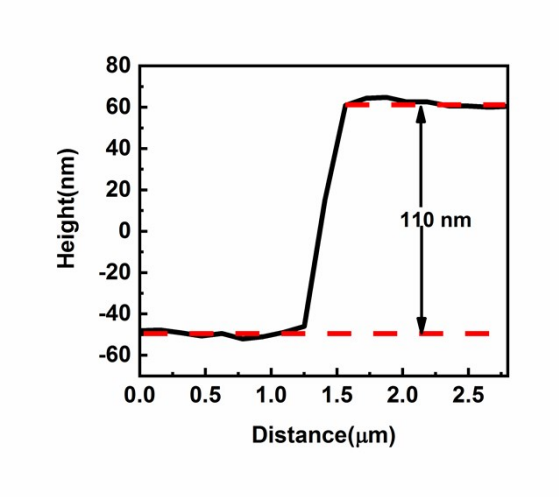


Fig. S5 AFM Height curve of the device in Fig.5d.

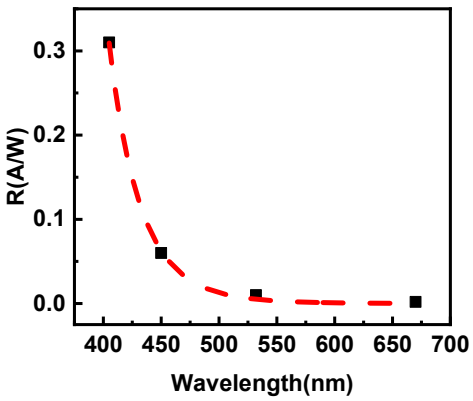


Fig. S6 Wavelength-dependent responsivity of the device with 14 nm.

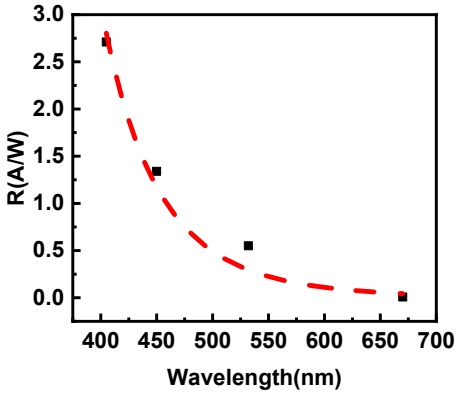


Fig. S7 Wavelength-dependent responsivity of the device with 110 nm.

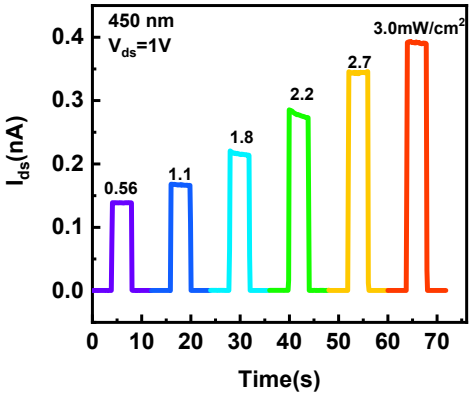


Fig. S8 I-t Curves of 450 nm with different power density

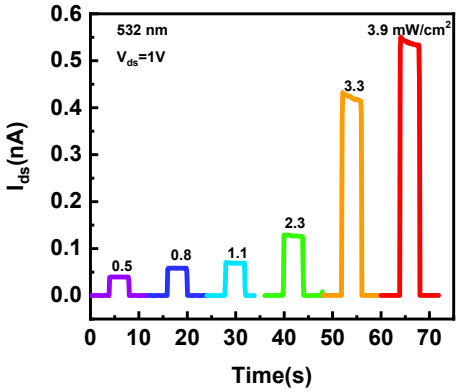


Fig. S9 I-t Curves of 532 nm with different power density

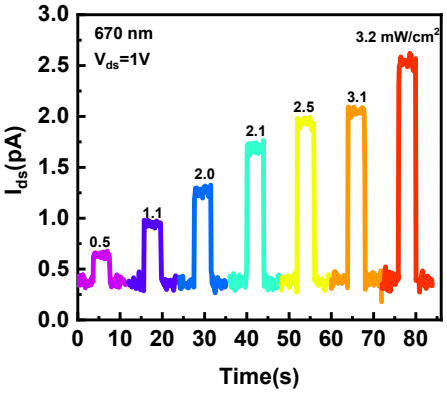


Fig. S10 I-t Curves of 670 nm with different power density

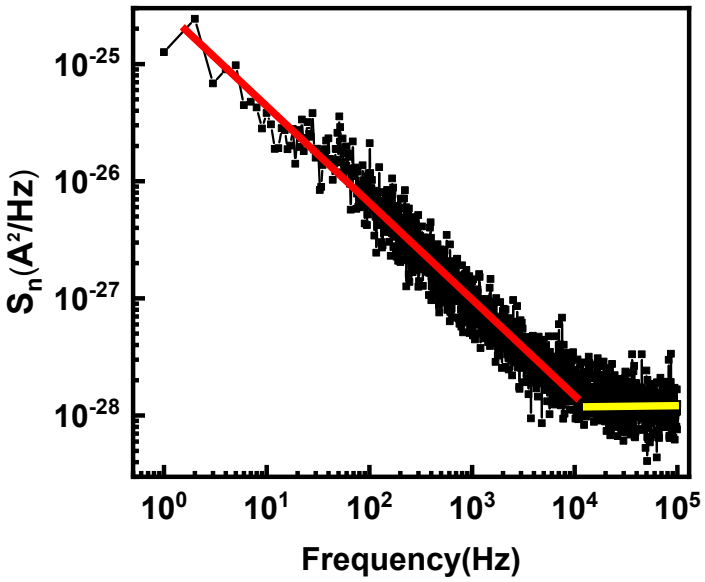


Fig. S11 The current noise power spectra of the device at V_{ds}=1 V

Table S1 The wavelength-dependent photoresponse for $In_{4/3}P_2Se_6$ flakes

Thickness (nm)	Wavelength (nm)	Responsivity (A/W)	Detectivity (Jones)	EQE (%)
14nm	405	0.31	3.63×10^{12}	94.9
	450	0.06	7.02×10^{11}	16.5
	532	0.01	1.17×10^{11}	2.33
	670	0.002	2.34×10^{10}	0.37
26nm	405	0.4	4.02×10^{12}	122
	450	0.22	1.12×10^{12}	60.6
	532	0.17	8.67×10^{11}	39.6
	670	0.011	5.61×10^{11}	2.04
110nm	405	2.71	2.47×10^{13}	829
	450	1.56	1.42×10^{13}	430
	532	0.55	5.05×10^{12}	102
	670	0.008	7.2×10^{10}	1.5

Table S2 The Detectivity (D_n) Comparision of 2D materials Based Photodetectors

2D Materials	Detectivity(Jones)	Ref.
$In_{4/3}P_2Se_6$	1.7×10^{11} 1.2×10^{12}	This work
NbS_3	5.7×10^{8}	S1
BP	5.2×10^{10}	S2
Fe_3O_4	7.42×10^{8}	S3
Se_xTe_{1-x}	6.5×10^{10}	S4
SnS	6.83×10^{10}	S5
$InAs_{0.40}P_{0.60}$	2×10^{10}	S6
BiOBr	1.44×10^{11}	S7

The calculation process of Planes angle

For hexagonal system, the angle between the crystal planes can be calculated according to the following formula.

$$\cos\phi = \frac{h_1h_2 + k_1k_2 + \frac{1}{2}(h_1k_2 + h_2k_1) + \frac{3a^2}{4c^2}l_1l_2}{\sqrt{(h_1^2 + k_1^2 + h_1k_1 + \frac{3a^2}{4c^2}l_1^2)(h_2^2 + k_2^2 + h_2k_2 + \frac{3a^2}{4c^2}l_2^2)}}$$

The angle between planes (300) and (3**Error!**0) is

$$\cos\phi = \frac{3\times3 + 0\times(-3) + \frac{1}{2}[3\times(-3) + 3\times0]}{\sqrt{3^2\times[3^2 + 3^2 + (-3)\times3)}}$$

$$= \frac{1}{2}$$

So ϕ=60°.

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