

Supplementary Information

High-responsivity Two-dimensional p-PbI₂/n-WS₂ Vertical Heterostructure Photodetectors

Enhanced by Photogating Effect

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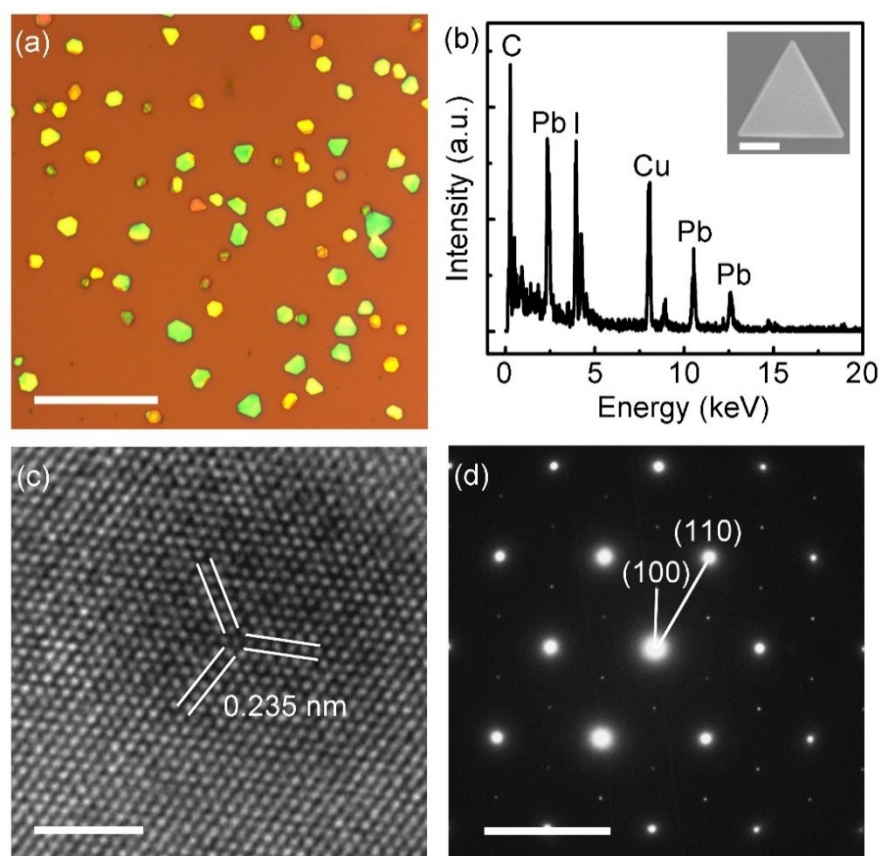


Fig. S1 (a) Optical image of as-grown PbI₂ nanoplates. Scale bar, 20 μ m. (b) EDS spectrum collected from a representative nanoplate. Insert is the corresponding SEM image. Scale bar, 200 nm. (c) HRTEM image of PbI₂ nanoplate. The lattice spacings along three different

directions are 0.235 nm, with an intersection angle of 60° . Scale bar, 2 nm. (d) SAED pattern viewed along [001] zone axis. Scale bar, 2 nm^{-1} .

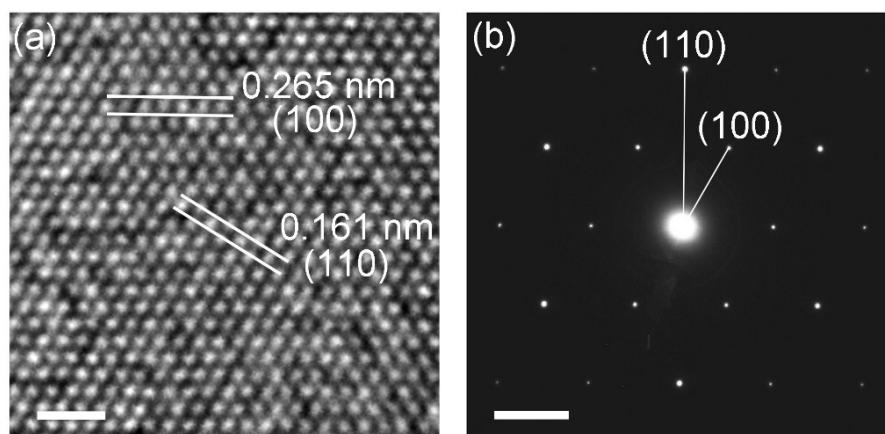


Fig. S2 (a) HRTEM image of monolayer WS_2 transferred on copper grid. (b) SAED pattern viewed along the [001] zone axis. Scale bars, 2 nm, 3 nm^{-1} , respectively.

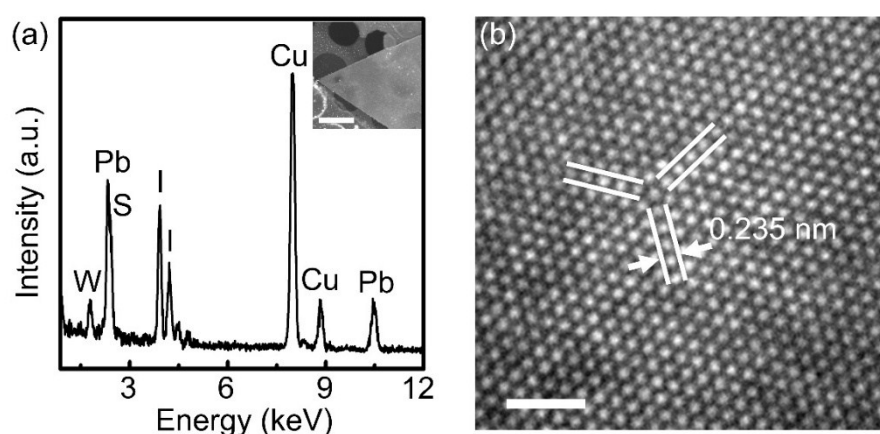


Fig. S3 (a) EDS result of PbI_2/WS_2 nanoplate on copper grid. Inset: low-resolution TEM image. Scale bar: $5 \mu\text{m}$. (b) HRTEM image of a typical nanoplate. The interplanar spacings along three directions (marked by yellow lines) are 0.235 nm with an intersection angle of 120° , corresponding to the {110} crystal plane family of PbI_2 . Scale bar: 1 nm.

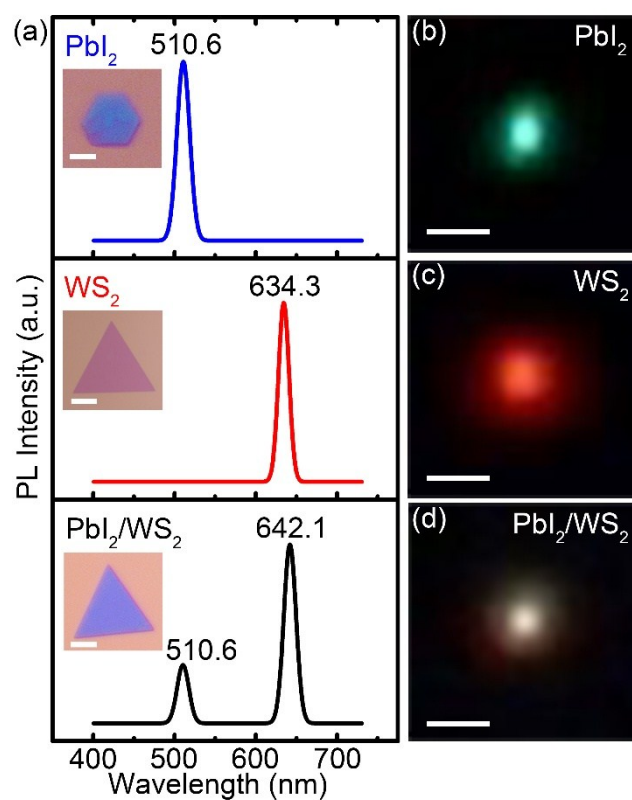


Fig. S4 (a) PL spectra of pristine Pbl₂ nanoplate, monolayer WS₂ and Pbl₂/WS₂ heterostructure, excited by 450 nm laser. Insert is the corresponding optical images of the measured samples. Scale bars, 2 μm , 5 μm and 5 μm . The real-color PL images are demonstrated in (b-d), respectively. Scale bars, 1 μm .

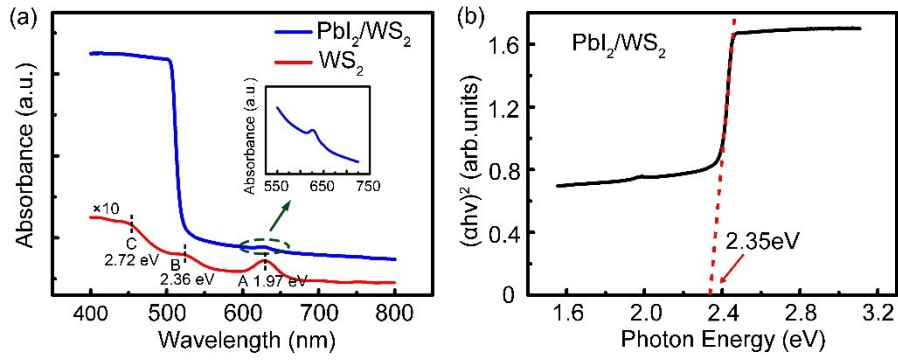


Fig. S5 (a) UV-vis absorption spectra of PbI_2/WS_2 heterostructure and pristine WS_2 (the absorbance intensity of WS_2 is multiplied by 10). The three peaks in the WS_2 absorption spectrum are assigned to A (1.97 eV), B (2.36 eV) and C (2.72 eV) excitonic absorption of pristine WS_2 . Insert is the amplification region from PbI_2/WS_2 absorption spectrum. (b) Tauc plot of PbI_2/WS_2 nanoplates absorption spectrum. Bandgap of PbI_2 was deduced to be 2.35 eV from absorbance data using the Tauc relation.^[1-3]

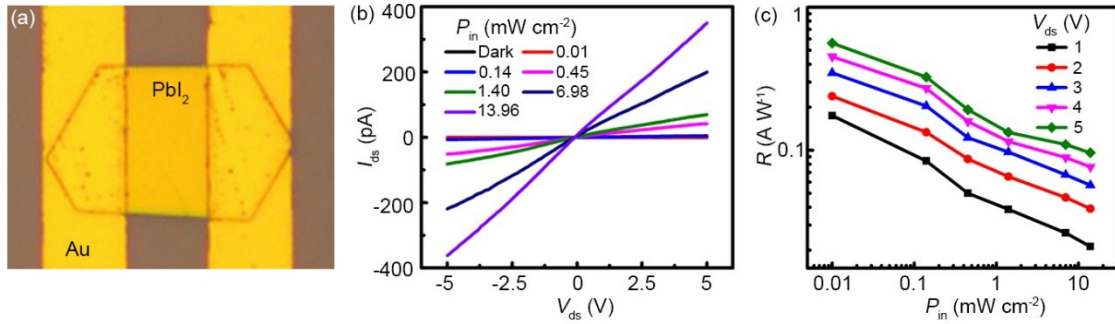


Fig. S6 Optoelectronic characterizations of pristine PbI_2 nanoplatform. (a) Optical image of a fabricated PbI_2 device on SiO_2/Si substrate. Scale bar, 5 μm . (b) $I_{\text{ds}} - V_{\text{ds}}$ curves of the device measured in dark and under various illumination power densities of 450 nm laser ($V_{\text{gs}} = 0 \text{ V}$). (c) Calculated photoresponsivity as a function of power density at different source-drain voltages.

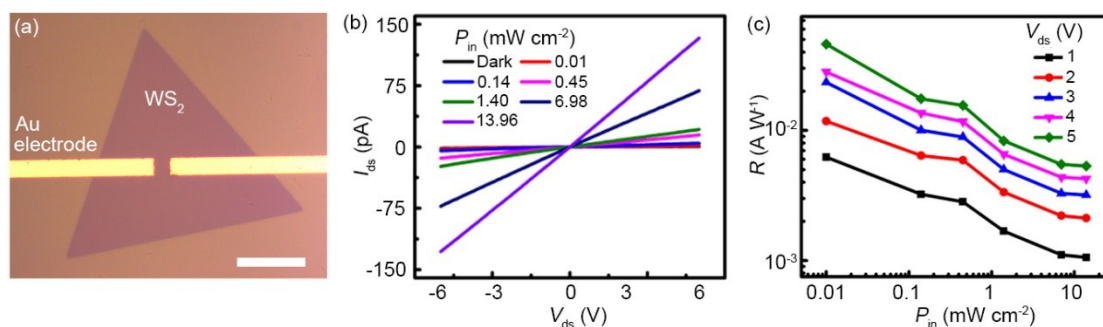


Fig. S7 Optoelectronic performance of monolayer WS₂ nanosheet. (a) Optical image of a representative WS₂ device on SiO₂/Si substrate. Scale bar, 20 μm. (b) I_{ds} - V_{ds} curves of the photodetector measured in dark and under various illumination power densities of 450 nm laser ($V_{gs} = 0$ V). (c) Extracted photoresponsivity as a function of illumination power density at different source-drain voltages.

Table S1: Comparison with vertical p-n heterostructure and pristine material photodetectors synthesized by CVD method.

Materials	Wavelength [nm]	V_{ds} [V]	V_{gs} [V]	Responsivity [$A W^{-1}$]	Detectivity [Jones]	Reference
SnS/WS ₂	532	1	0	24.2	-	[4]
GaSe/MoSe ₂	white light	5	0	0.03	-	[5]
WSe ₂ /SnS ₂	520	5	0	0.11	4.71×10^{10}	[6]
GaSe/MoS ₂	300	1	0	~3.75	-	[7]
BiI ₃ /WSe ₂	white light	1	0	1.67	-	[8]
ML ^{a)} WS ₂	white	20	60	1.88×10^{-2}	-	[9]
FL ^{b)} PbI ₂	450	1.9	0	1×10^{-4}	-	[10]
ML WS ₂	450	5	0	5.0×10^{-2}	-	This work
FL PbI ₂	450	5	0	5.6×10^{-1}	-	This work
PbI ₂ /WS ₂	450	5	0	5.57×10^2	6.8×10^{12}	This work
PbI ₂ /WS ₂	450	5	-60	7.1×10^4	3.7×10^{13}	This work

^{a)} ML: monolayer; ^{b)} FL: fewlayer

REFERENCES

- 1 B. Saparov, F. Hong, J. Sun, H. Duan, W. Meng, S. Cameron, I. G. Hill, Y. Yan and D. B. Mitzi, *Chem. Mater.*, 2015, **27**, 5622-5632.
- 2 P. Cheng, T. Wu, Y. Li, L. Jiang, W. Deng and K. Han, *New J. Chem.*, 2017, **41**, 9598-9601.
- 3 M. Leng, Y. Yang, K. Zeng, Z. Chen, Z. Tan, S. Li, J. Li, B. Xu, D. Li, M. P. Hautzinger, Y. Fu, T. Zhai, L. Xu, G. Niu, S. Jin and J. Tang, *Adv. Funct. Mater.*, 2018, **28**, 1704446.
- 4 A. Sukma Aji, M. Izumoto, K. Suenaga, K. Yamamoto, H. Nakashima and H. Ago, *Phys. Chem. Chem. Phys.*, 2018, **20**, 889-897.
- 5 X. Li, M.-W. Lin, J. Lin, B. Huang, A. A. Puretzky, C. Ma, K. Wang, W. Zhou, S. T. Pantelides and M. Chi, *Sci. Adv.*, 2016, **2**, e1501882.
- 6 T. Yang, B. Zheng, Z. Wang, T. Xu, C. Pan, J. Zou, X. Zhang, Z. Qi, H. Liu, Y. Feng, W. Hu, F. Miao, L. Sun, X. Duan and A. Pan, *Nat. Commun.*, 2017, **8**, 1906.
- 7 N. Zhou, R. Wang, X. Zhou, H. Song, X. Xiong, Y. Ding, J. Lu, L. Gan and T. Zhai, *Small*, 2018, **14**.
- 8 J. Li, X. Guan, C. Wang, H. C. Cheng, R. Ai, K. Yao, P. Chen, Z. Zhang, X. Duan and X. Duan, *Small*, 2017, **13**.
- 9 C. Lan, C. Li, Y. Yin and Y. Liu, *Nanoscale*, 2015, **7**, 5974-5980.
- 10 W. Zheng, Z. Zhang, R. Lin, K. Xu, J. He and F. Huang, *Adv. Electron. Mater.*, 2016, **2**, 1600291.