Supporting Information for

Out of plane stacking InSe-based heterostructures towards high performance electronic and optoelectronic devices using graphene electrode

Wei Gao[1], Zhaoqiang Zheng[1*], Yongtao Li[1], Congxin Xia[2], Juan Du[2], Yu Zhao[1], Jingbo Li[1,3,4*]

[1] College of Materials and Energy, Guangdong University of Technology, Guangzhou 510006, P. R. China.

[2] Physics and Electronic Engineering College, Henan Normal University, Xinxiang 453007, P. R. China.

[3] State Key Laboratory of Precision Electronics Manufacturing Technology and Equipment, Guangdong University of Technology, Guangzhou 510006, P. R. China.

[4] State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, P. R. China.

[*] Corresponding authors: zhengzhq5@mail2.sysu.edu.cn; jbli@semi.ac.cn

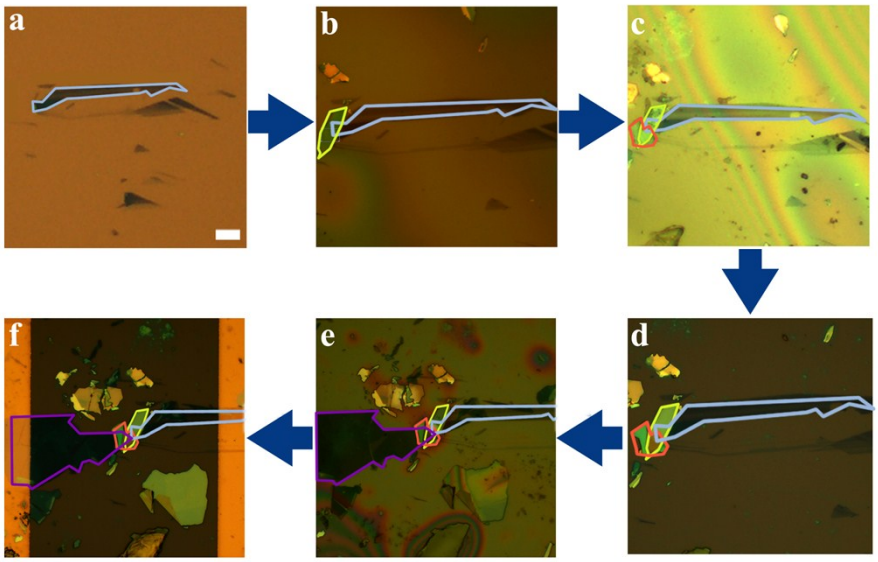

Fig. S1 Fabrication process of the Gr-InSe/WSe_2-Gr heterostructure device. (a) Exfoliation of bottom graphene on SiO_2/Si substrate. The scale bar is 10 μm. (b) Stacking of WSe_2. (c) Stacking of InSe with PMMA film. (d) Remove PMMA. (e) Stacking of top graphene. (f) Patterning metal contacts to top-bottom graphene.

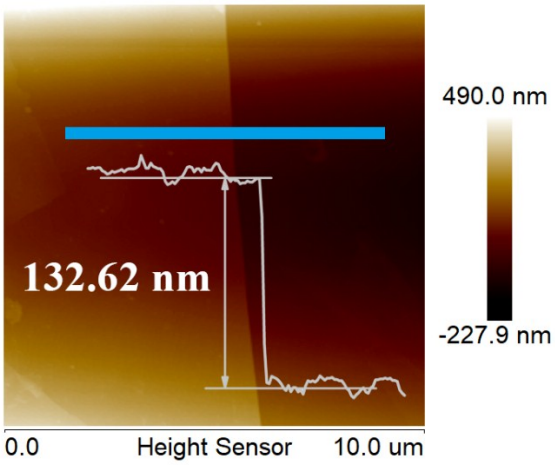

Fig. S2 Atomic Force microscopy (AFM) topography at the interface of the top graphene.

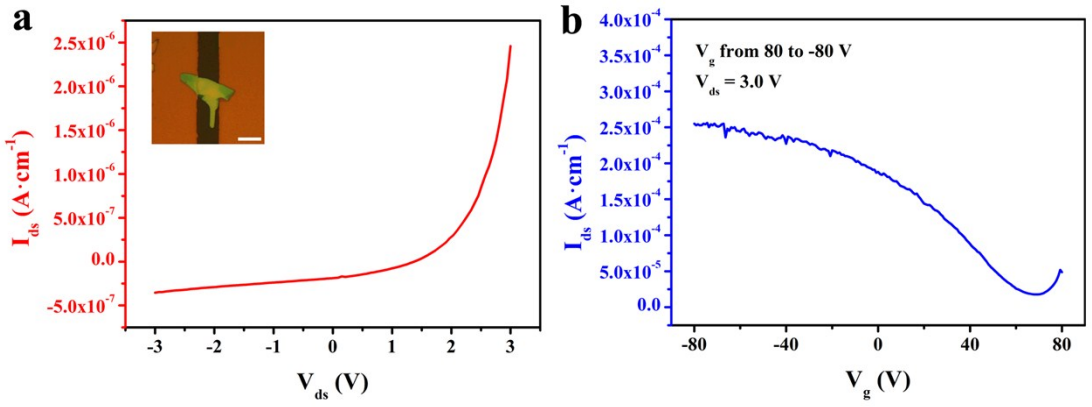

Fig. S3 Electrical performance of the Au-InSe-Au heterostructure device. (a) I_{ds}-V_{ds} curve of the device under dark condition. Inset is the optical image of the FETs device showing the red overlapped region, the scale bar is 10 μm. (b) Transfer curves as a function of gate voltage under dark condition.

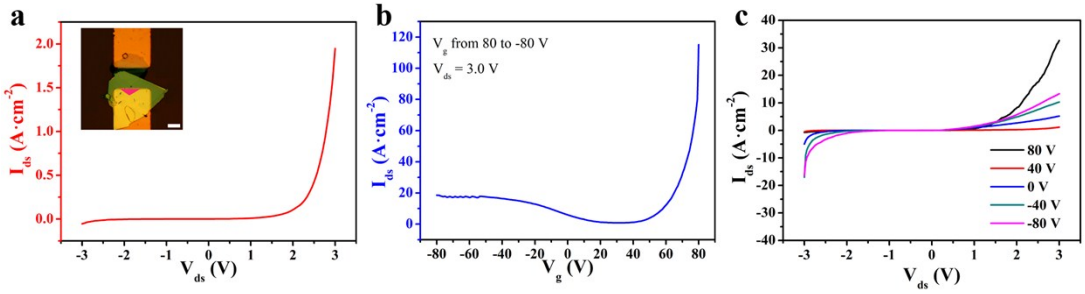

Fig. S4 Electrical performance of the Au-WSe_2-Gr heterostructure device. (a) I_{ds}-V_{ds} curve of the device under dark condition. Inset is the optical image of the FETs device showing the red overlapped region, the scale bar is 10 μm. (b) Transfer curves as a function of gate voltage under dark condition. (c) Output characteristic curves of the device with gate voltage from 80 V to -80 V.

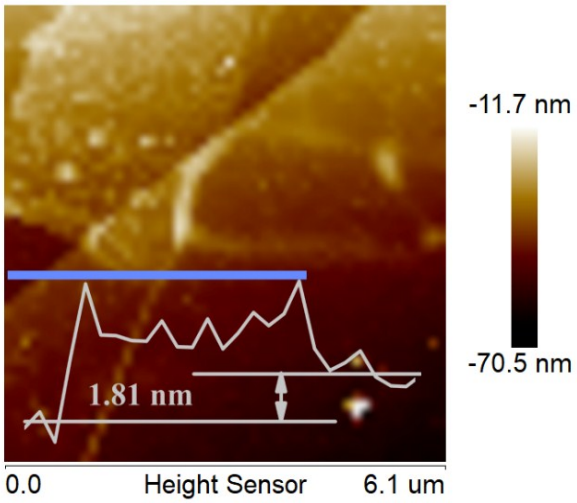

Fig. S5 Atomic Force microscopy (AFM) topography at the interface of the top graphene.

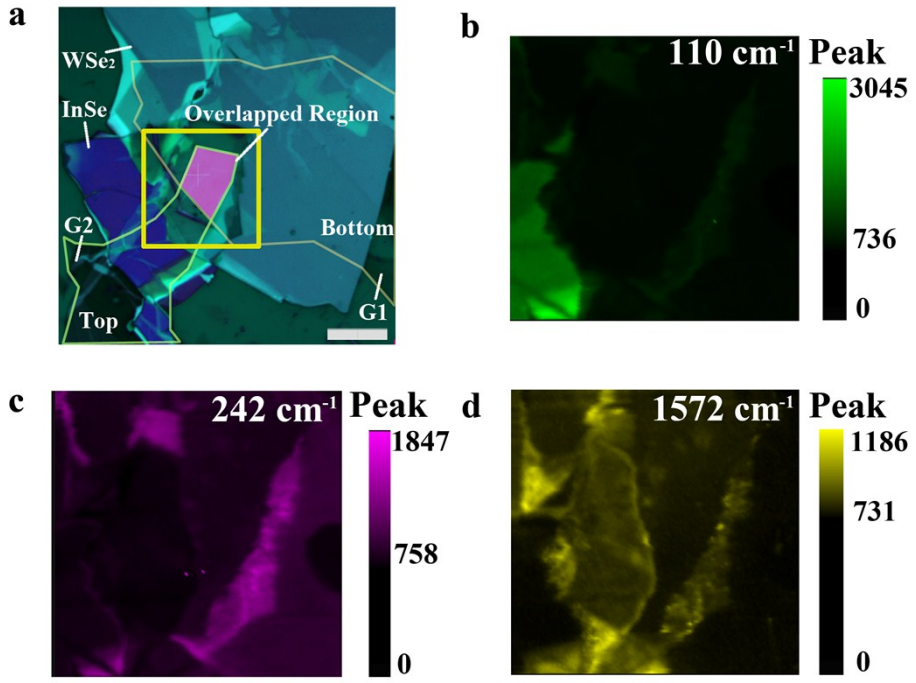

Fig. S6 (a) Optical image of the heterostructure with larger overlapped region. The Raman mapping of the yellow rectangle in (a): (b) 110 cm^{-1} (InSe); (c) 242 cm^{-1} (WSe_2); (d) 1572 cm^{-1} (Graphene). The scale bar is 10 μm. The overlapped region is about 159 μm^2.

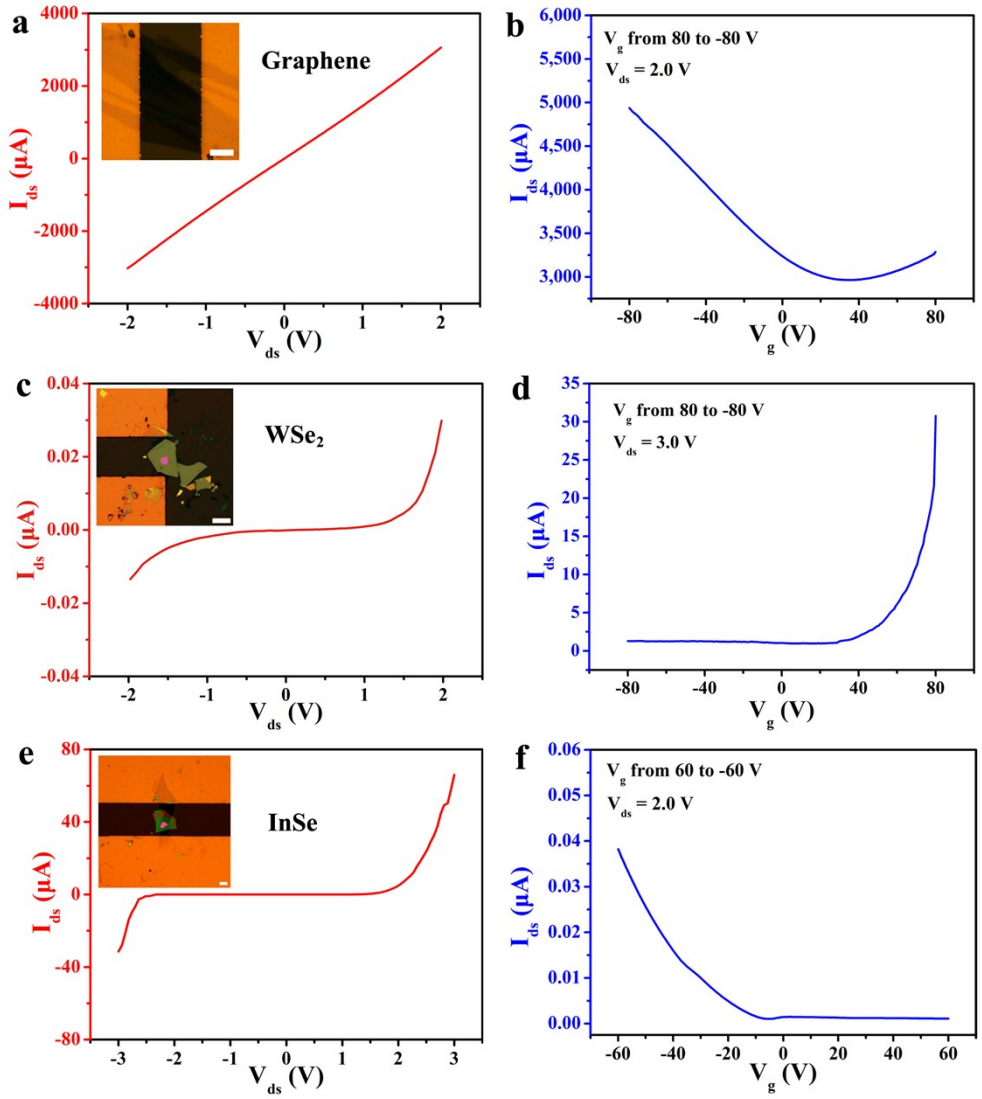

Fig. S7 Electrical properties of the FETs devices for graphene, vertical Gr-WSe_2-Gr and Gr-InSe-Gr. (a), (c), (e) I_{ds}-V_{ds} characteristic curves without the gate voltage. (b), (d), (f) Transfer curves. All scale bars are 10 μm. The pink regions are overlapped areas.

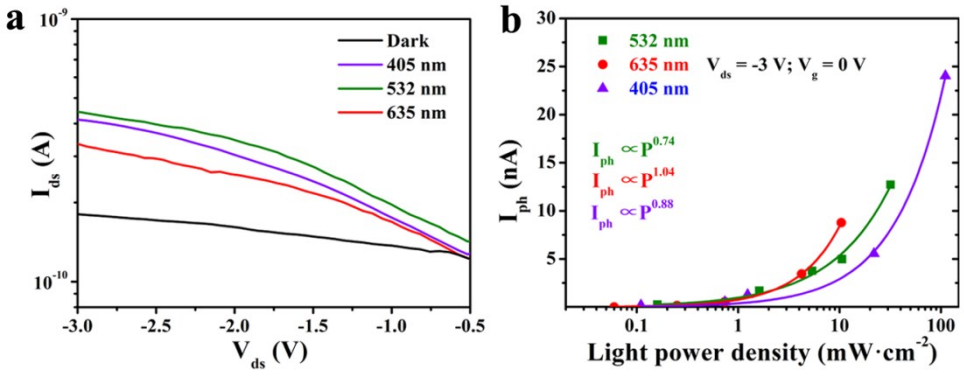

Fig. S8 (a) I_{ds}-V_{ds} of the Gr-InSe/WSe_2-Gr photodetector under 405 nm, 532 nm and 635 nm or without illumination, the light power density is 0.27 mW·cm^{-2}, 0.16 mW·cm^{-2} and 0.25 mW·cm^{-2}; respectively. (b) Light power density dependence of the photocurrent.

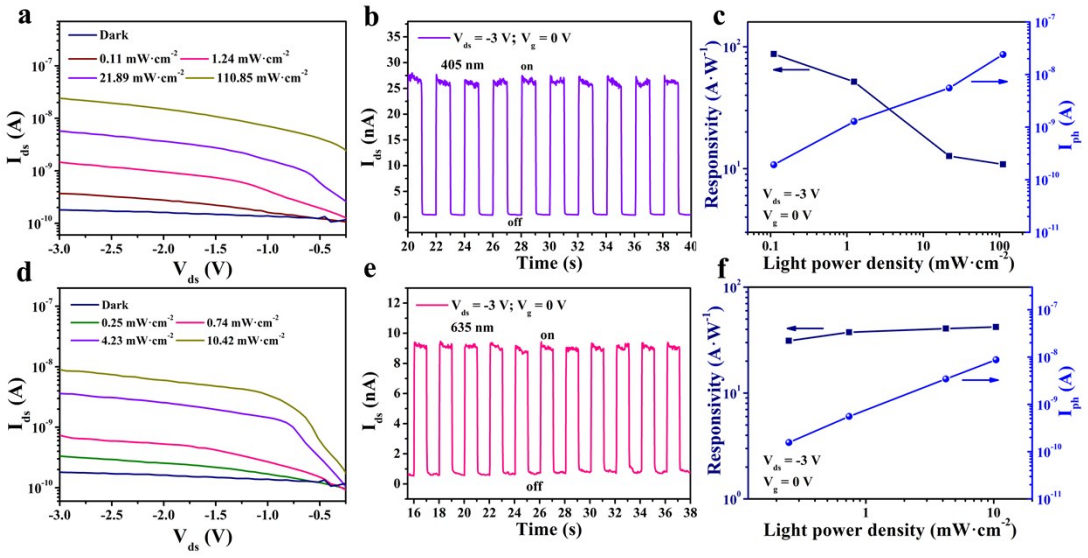

Fig. S9 Optoelectrical properties of the Gr-InSe/WSe_2-Gr. (a) I_{ds}-V_{ds} curves dependent on the light power density under a 405 nm illumination and at dark. (b) Time trace of I_{ds} under a 405 nm illumination (110 mW·cm^{-2}). (c) Photoresponsivity and photocurrent as a function of light power density under 405 nm. (d) I_{ds}-V_{ds} curves dependent on the light power density under a 635 nm illumination and at dark. (e) Time trace of I_{ds} under a 635 nm illumination (10.42 mW·cm^{-2}). (f) Photoresponsivity and photocurrent as a function of light power density under 635 nm.

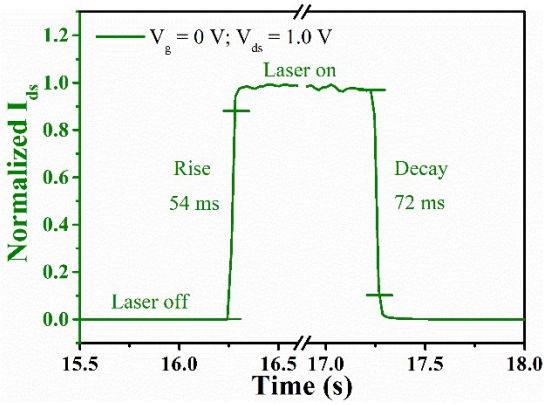

Fig. S10 Rising and decay time of the Gr-InSe-Gr photodetector under a 532 nm illumination (P_{laser} = 238.9 mW·cm^{-2}).

Table S1 Comparison of figures-of-merit for vertical graphene-photodetectors based on 2D layered materials

Materials	Measurement condition	Responsivity (A/W)	EQE (%)	D* (Jones)	Response time	Ref.
Graphene-WSe_2/ p-InSe-Graphene	532 nm V_g = 0 V V_{ds} = 3 V	83	1.93×10^4	1.55×10^{12}	36ms /18ms	In this work
Graphene-$MoTe_2$ -Graphene	1064 nm V_g = 30 V V_{ds} = 0 V	0.11	12.9	/	24μs	1
Graphene-n-InSe -Graphene	633 nm V_g = 0 V V_{ds} = 2 V	10^5	10^5	10^{13}	/	2
Graphene-MoS_2 -Graphene	514 nm V_g = -60 V V_{ds} = 0.5 V	/	25	/	50μs	3
Graphene-p-GaSe/ n-InSe-Graphene	410 nm V_g = 0 V V_{ds} = 2 V	350	/	3.7×10^{12}	2μs	4
Graphene-Ta_2O_5 -Graphene	532 nm V_{ds} = 1 V	10^3	/	/	0.75s	5
Graphene-WSe_2/ GaSe-Graphene	520 nm V_g = 0 V V_{ds} = -1.5 V	6.2±0.2	1490± 50	/	30μs	6
Graphene-WSe_2 -Graphene	759 nm V_g = 0 V V_{ds} = 0.5 V	/	7.3	/	1.6ns	7
h-BN-Graphene-MoS_2/WSe_2-Graphene	532 nm V_g = 0 V V_{ds} = 0 V	0.12	34	/	/	8

Table S2 Comparison of figures-of-merit for lateral graphene-photodetectors based on 2D materials

Materials	Measurement condition	Responsivity (A/W)	EQE (%)	D* (Jones)	Response time	Ref.
Graphene-WSe_2 /p-InSe-Graphene	532 nm V_g = 0 V V_{ds} = 3 V	83	1.93×10^4	1.55×10^{12}	36ms/18ms	In this work
Graphene-n-InSe -Graphene	633 nm V_g = 0 V V_{ds} = 2 V	4×10^3	5×10^3	10^{10}	0.1 ms	2
Graphene-n-InSe-Graphene	500 nm V_g = 0 V V_{ds} = 10 V	60	14850	/	120μs+5s	9
MoS_2-Glassy-Graphene	532 nm V_g = 0 V V_{ds} = 1 V	0.0123	/	1.8×10^{10}	35s	10
Graphene-WS_2/MoS_2-Graphene	532 nm V_g = 0 V V_{ds} = 10 V	2340	5460	4.1×10^{11}	>1s	11
Graphene-WS_2-Graphene	532 nm V_g = 80 V V_{ds} = 5 V	4500	/	/	2ms	12

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