Electronic Supplementary Information

Cation Exchange Synthesis of Two-dimensional Vertical Cu_2S/CdS Heterojunctions for Photovoltaic Device Applications

Yangyang Zhan,[a] Zhibin Shao,[a] Tianhao Jiang,[a] Jing Ye,[a] Xiaofeng Wu,[a] Bingchang Zhang,[a] Ke Ding,[a] Di Wu,[b] and Jiansheng Jie*[a]*

[a] Institute of Functional Nano & Soft Materials (FUNSOM), Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Testing & Analysis Center, Soochow University, Suzhou, Jiangsu 215123, P. R. China

[b] School of Physics and Engineering, Key Laboratory of Material Physics of Ministry of Education, Zhengzhou University, Zhengzhou, Henan 450052, P. R. China

Email: jsjie@suda.edu.cn; zbshao@suda.edu.cn.

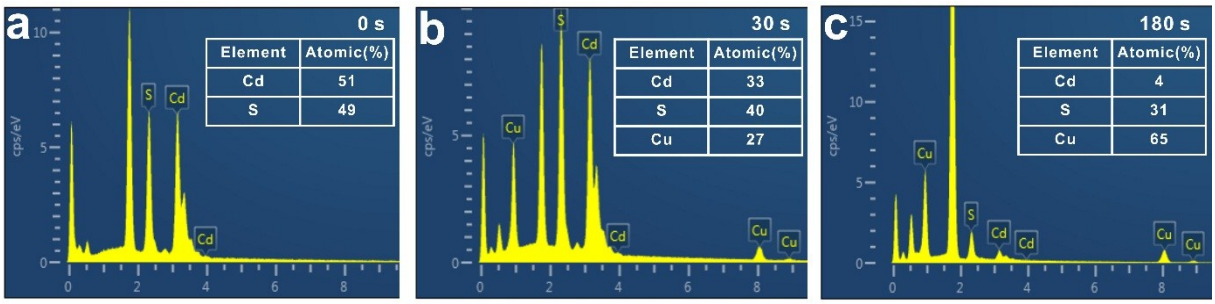

Fig. S1 EDS spectra of the nanosheet at different reaction times of (a) 0, (b) 30, and (c) 180 s.

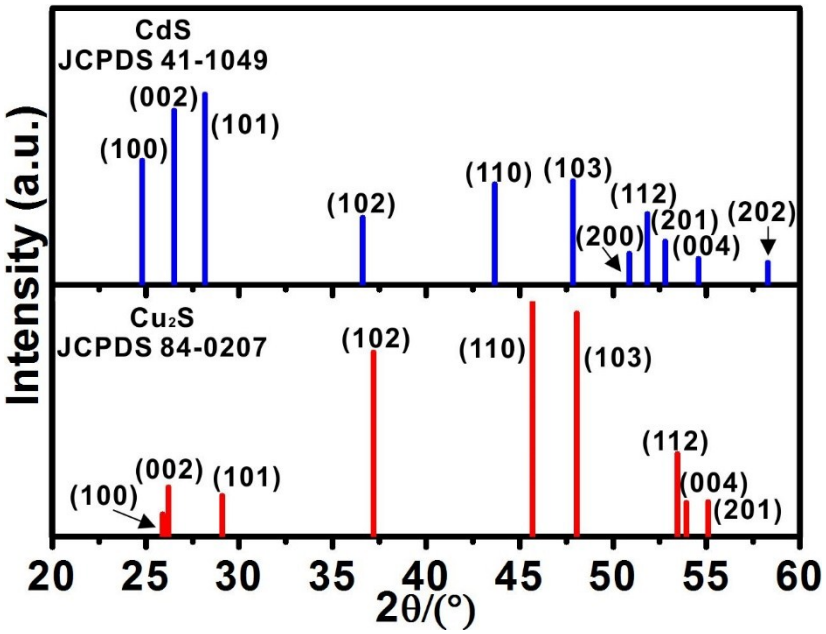

Fig. S2 XRD spectra of the hexagonal CdS (JCPDS #41-1049) and Cu_2S (JCPDS #84-0207).

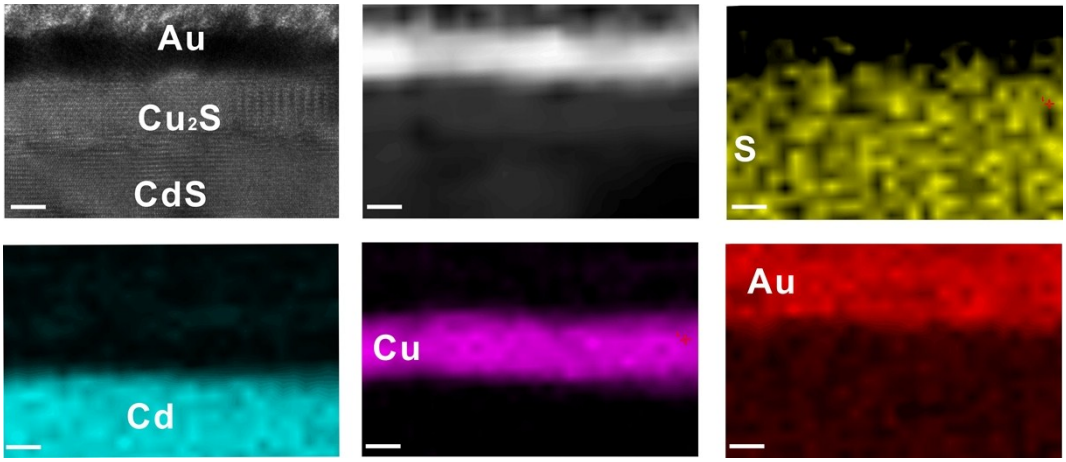

Fig. S3 Cross-sectional TEM, STEM and the corresponding EDS element mapping images of the vertical Cu_2S/CdS heterostructure in the z-axis direction. EDS element mapping includes the S (yellow), Cd (cyan), Cu (purple), and Au (red) element. In the cross-section TEM characterization, an Au film was deposited on the upper surface of nanosheet to improve conductivity. The scale bars represent 5 nm.

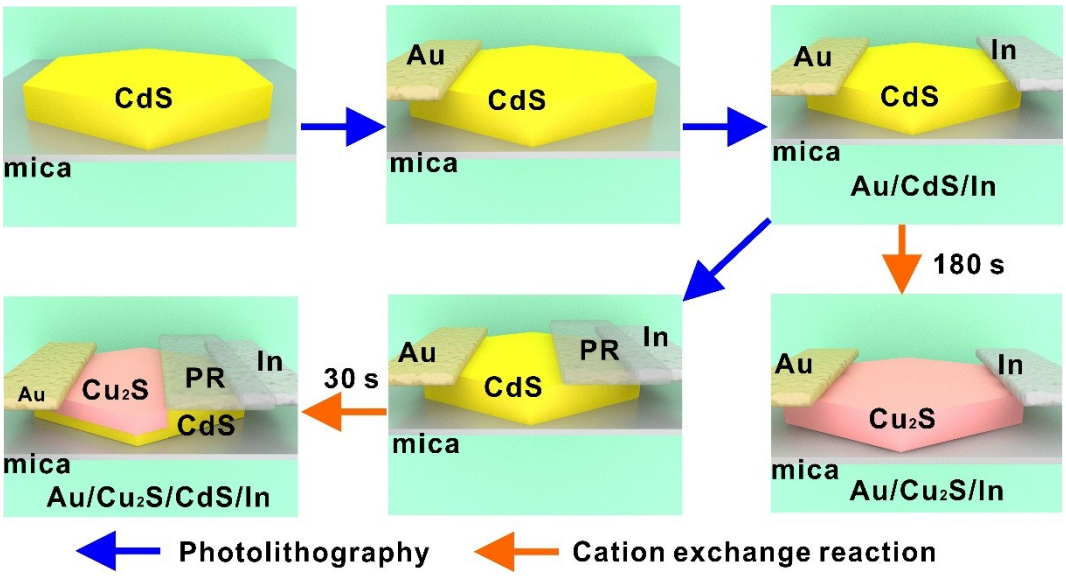

Fig. S4 Schematic illustration of the fabrication process for the Au/Cu_2S/CdS/In-based photovoltaic device.

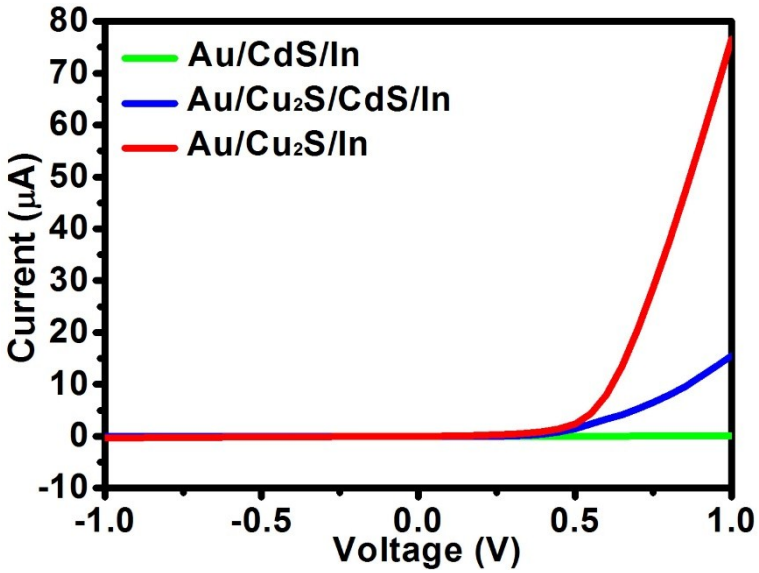

Fig. S5 *I-V* curves of the devices measured under light irradiation of 0.3 mW cm^{-2}.

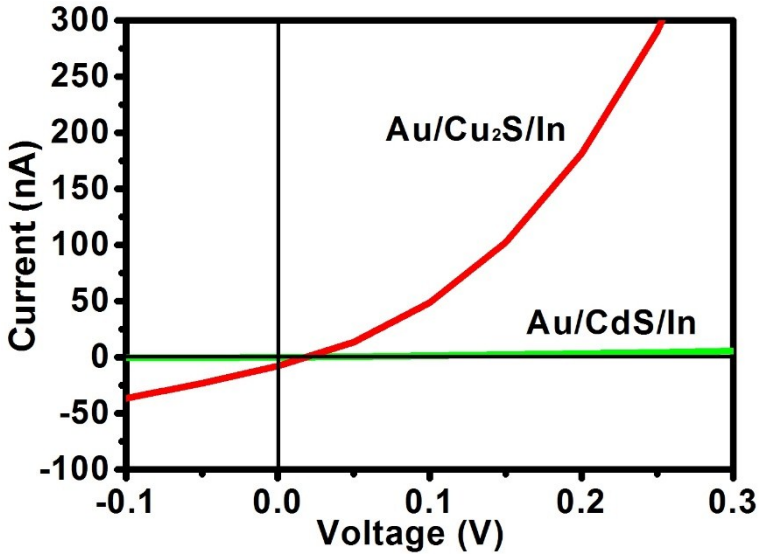

Fig. S6 Enlarged *I-V* curves of the devices based on Cu_2S and CdS nanosheets.

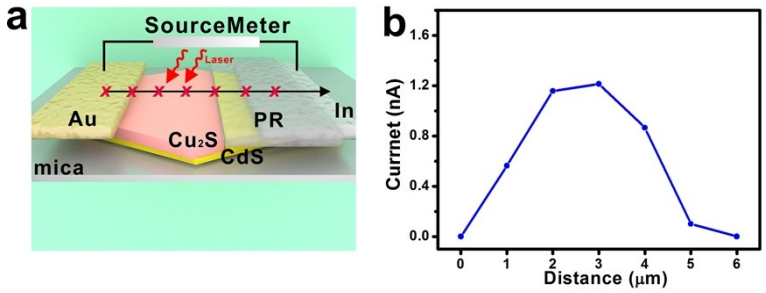

Fig. S7 (a) Schematic illustration and (b) photocurrent line-scanning profile of a scanning photocurrent measurement. The device was irradiated point by point by a laser beam with a spot size of less than 2 μm along the direction of the arrow, while the photocurrent of the device was recorded by applying a zero bias.

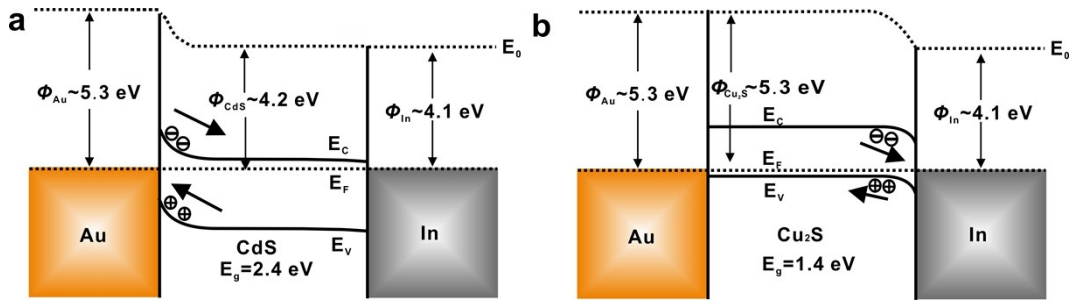

Fig. S8 Energy band diagrams of (a) Au/CdS/In and (b) Au/Cu_2S/In Schottky junction devices.

Table S1 Performance comparison of 2D Cu_2S/CdS heterojunctions with other lateral and vertical heterojunctions based on 2D layered materials.

Materials	Types	Methods	V_{OC} (V)	I_{SC}/J_{SC}	*FF*	PCE (%)	Power	Ref.
CdS/Cu_2S	Vertical junctions	Cation exchange reaction	0.17	2.3 nA	0.483	2.1	0.3 mW cm^{-2}	This work
WSe_2/MoS_2	Lateral junctions	CVD	0.22	0.008 nA	0.39	0.2	1 mW cm^{-2}	1
WS_2/WSe_2	Lateral junctions	CVD	0.47	0.2 nA		0.9	30 nW	2
MoS_2/WS_2	Lateral junctions	CVD	0.32	3.5 pA	0.27		12.63 mW cm^{-2}	3
WSe_2/MoS_2	Lateral junctions	CVD	0.39	0.71 nA	0.37	2.56	100 mW cm^{-2}	4
WSe_2/MoS_2	Vertical junctions	Mechanical exfoliation			~0.5	0.2	640 mW cm^{-2}	5
GaTe/InSe	Vertical junctions	Mechanical exfoliation			0.25	0.1	6.35 mW cm^{-2}	6
BP/WS_2	Vertical junctions	Mechanical exfoliation			0.33	4.6	36 mW cm^{-2}	7
WSe_2/MoS_2	Vertical junctions	Dry transfer				0.4	100 mW cm^{-2}	8
MoS_2/AsP	Vertical junctions	Dry transfer	0.61	1.3 μA	0.5	9	108.8 mW cm^{-2}	9
GaSe/$MoSe_2$	Vertical junctions	CVD	0.57	0.35 mA cm^{-2}	0.38	0.12	64.42 mW cm^{-2}	10

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