

Supporting Information

High-performance hierarchical O-SnS/I-ZnIn₂S₄ photodetectors by leveraging the synergy of optical regulation and band tailoring

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S1. Experimental section

Cleaning of Substrates: Before the PLD deposition of materials, ultrasonic treatment is used to remove the remaining contamination on the surface of the SiO₂/Si, Si and sapphire substrates by sequentially immersing them into acetone, ethanol and deionized water. Each cleaning step takes about 15 minutes.

Synthesis of ZIS Nanofilms: High-quality ZIS nanofilms are obtained by PLD deposition and post-growth annealing treatment.

PLD Deposition: A KrF excimer laser (COMPex Pro 110F, Coherent GmbH, Göttingen, Germany, $\lambda = 248$ nm) is used for ablating the ZIS target. The pulse number for depositing the ZIS nanofilms is set to be 5000 with the repetition rate of 4 Hz and the operating voltage of 19 kV. The distance between the target and substrate during deposition is approximately 6 cm and the temperature of the substrate holder is set to be 450 °C during the ZIS deposition process. The base pressure of the growth chamber is less than 6×10^{-4} Pa, and the growth pressure is set at 40 Pa through aerating 99.99% argon gas at a flow rate of 50 sccm.

Post-Growth Annealing Process: After the PLD deposition, the derived ZIS nanofilms are transferred into a tube furnace (GSL-1100X-S) for annealing treatment. The ZIS nanofilms are set at the middle of the heating zone of the furnace, and the sulfur powder (600 mg, $\geq 99.95\%$, Aladdin) is placed on the upstream side of the furnace. To provide a protective gas environment, the tube is flushed with flowing high-purity argon (99.99%) gas for 40 minutes before heating. Then, the furnace temperature is increased to 600 °C with a heating rate of 10 °C/min and is kept at 600 °C for 30 minutes. With the assist of the flowing high-purity argon gas, the gasified sulfur species are supplied to the ZIS nanofilms to create a sulfur-rich environment in the furnace.

Synthesis of the SnS/ZIS Heterojunctions: The SnS nanofilms are deposited over the ZIS nanofilms which are obtained by the above two-step process to produce the SnS/ZIS nanofilms. The target is consisted of highly pure SnS₂ powder (99.99%), which is prepared by a sintering process. The pulse number for depositing the SnS nanofilms is set to be 1000, 2000, 3000 and 4000 with the repetition rate of 4 Hz and the operating voltage of 19 kV. The distance between the target and substrate during the PLD deposition is approximately 6 cm, and the temperature of substrate holder is set at

500 °C during the deposition process. The base pressure of the growth chamber is less than 6×10^{-4} Pa and the growth pressure is set at 40 Pa through aerating 99.99% argon gas at a flow rate of 50 sccm.

Material Characterizations: The crystal structure of the ZIS, SnS and SnS/ZIS nanofilms are recorded by an XRD system (Rigaku D-MAX 2200 VPC). Raman spectroscopy and PL spectrum measurements are performed at room temperature by a Renishaw InVia spectrometer with an excitation wavelength of 514 nm. The morphology characterizations of the ZIS, SnS and SnS/ZIS nanofilms are achieved by SEM (JSM-6330F). TEM (FEI Tecnai G2 F30) is used to perform further structure and morphology characterizations. The components and structure of the nanofilms are extracted by energy dispersive spectroscopy (EDS) and selected area electron diffraction (SAED) measurements. XPS is recorded with a Thermo Scientific XPS system (Nexsa). The UV-Vis-NIR absorption spectra and reflectance spectra are measured with a UV-Vis-NIR spectrophotometer (Lambda950, Perkin Elmer). An atomic force microscopy system (AFM, Bruker Dimension FastScan) is used to conduct the roughness and thickness measurements.

Device Fabrication: The Ti/Au (10/100 nm) electrode array are patterned onto the nanofilms by photolithography (μ PG 501, Heidelberg) and electron beam evaporation (Wavetest, DE400).

Device Characterizations: The measurements of the electrical and photoelectric properties of the devices are carried out on a Lakeshore probe station combined with a semiconductor parameter analyzer (4200-SCS, Keithley). The photoresponse of the photodetectors is measured under 370.6 nm, 405 nm, 532 nm, 671 nm, 808 nm, 1550 nm, 2240 nm lasers in atmospheric environment at room temperature. A high-speed switch is used to generate the pulsed light. A Si photodiode (Throlabs S120VC, for $\lambda \leq 808$ nm) and a GaInAs photodiode (Throlabs S148C, for $\lambda \geq 1550$ nm) are exploited to calibrate the laser energy.

Theoretical Methods: The projector-augmented wave pseudopotentials (PAW) were used for relevant calculations, which were based on the Density Functional Theory (DFT) as implemented in the Vienna Ab-initio Simulation Package (VASP) software.^{1,2} The exchange-correlation potential was described by the Perdew-Burke-Ernzerhof (PBE)

in the generalized gradient approximation (GGA).^{3, 4} The kinetic energy cutoff for plane-wave expansion was set to 500 eV and the k-point grid in the Brillouin zones (BZ) of pure ZIS, SnS and SnS/ZIS heterojunction by executing Γ -centered $8 \times 5 \times 1$, $8 \times 8 \times 1$, and $8 \times 3 \times 1$,⁵ respectively. The full geometry relaxation until the absolute value of all forces was less than 0.01 eV/Å, and the energy convergence threshold for between two sequential steps is 10^{-5} eV. Moreover, The DFT-D3 correction was adopted to rectify the van der Waals interaction between ZIS and SnS layers. The vacuum area of ~ 25 Å was installed along the z -direction to minimize the spurious interactions between the repeating images.

The electronic transport and photocurrent calculations were performed by using the non-equilibrium Green's function (NEGF) combined with DFT within the implemented Atomistix ToolKit (ATK).⁶ The energy cutoff of 150 Ry and a k-point mesh of $1 \times 1 \times 100$ were used.⁷ The I - V curves of ZIS and SnS/ZIS heterojunctions were calculated in terms of the Landauer–Büttiker model:

$$I_{\sigma}(V_b) = G_0 \int_{\mu_L}^{\mu_R} T_{\sigma}(E, V_b) [f_L(E) - f_R(E)] dE, \quad (1)$$

where G_0 stands for the quantum unit of conductance, σ is the spin index and $f_{l(r)}(E)$ is the Fermi distribution function, $T_{\sigma}(E, V_b)$ represents the transmission coefficient under energy E and bias voltage V_b . A schematic diagram of the electronic device is presented in Fig. S1

In the light of the law of luminous flux conservation,⁸ the light absorptance of SnS nanosheets (A_{abs}^{SnS}) and $ZnIn_2S_4$ (A_{abs}^{ZIS}) in tilted/horizontal heterostructures can be described as

$$\begin{cases} A_{abs}^{SnS} = \frac{1 - R_A}{1 - R_A R_B T_1^+ T_1^-} \left(1 - R_B T_1^+ T_1^- - (1 - R_B) T_1^+ \frac{1 - R_C T_2^+ T_2^- [1 - (1 - R_B) T_1^-]}{1 - R_B R_C T_2^+ T_2^-} \right) \\ A_{abs}^{ZIS} = \frac{(1 - R_A)(1 - R_B) T_1^+}{1 - R_A R_B T_1^+ T_1^-} \left(1 - \frac{(1 - R_B) R_C T_2^+ T_2^- + (1 - R_C) T_2^+}{1 - R_B R_C T_2^+ T_2^-} \right) \end{cases}, \quad (2)$$

where R_A , R_B , R_C are the reflection coefficients at the interfaces of air/SnS, SnS/ZIS and air/ZIS, respectively.

$$R_A = (n_{air} - n_{SnS}^*)^2 / (n_{air} + n_{SnS}^*)^2, \quad (3)$$

$$R_B = (n_{SnS} - n_{ZIS})^2 / (n_{SnS} + n_{ZIS})^2, \quad (4)$$

$$R_C = (n_{ZIS} - n_{air})^2 / (n_{ZIS} + n_{air})^2, \quad (5)$$

$$n_{SnS}^* = n_{air} + \frac{HD}{L(H \sin \theta + D \cos \theta)} (n_{SnS} - n_{air}), \quad (6)$$

n_{SnS}^* is the effective refractive index of nanosheet array, θ is the tilt angle of SnS nanosheets, H and D are the height of SnS and the thickness of ZIS, and L is the array periodicity of SnS nanosheets, i.e., the distance between two adjacent nanosheets, n_{air} , $n_{SnS}=1.25$ and $n_{ZIS}=2.50$ are the refractive index of air, SnS and ZIS.^{9, 10} T_1^+ (T_2^+) and T_1^- (T_2^-) are the fraction of downward flux transmitted to the rear surface of SnS nanosheets (ZIS) and that of light reflected from the rear that is transmitted to the front surface of SnS nanosheets (ZIS), respectively. Physically, the fraction of luminous flux is

$$\begin{cases} T_1^\pm = 1 - \Gamma [1 - \exp(-\alpha_{SnS}(\lambda)(H \sin \theta + D \cos \theta))] \\ T_2^\pm = \exp(-\alpha_{ZIS}(\lambda)D) \end{cases}, \quad (7)$$

where Γ is the ratio of effective-to-actual illumination area, it is expressed as

$$\Gamma = (H \cos \theta + D \sin \theta) / L \quad (8)$$

α_{SnS} and α_{ZIS} are the light absorption coefficients of SnS nanosheets and ZIS that can be calculated by Tauc's law

$$\alpha = A_\alpha^* (h\nu - E_g)^\Omega / h\nu, \quad (9)$$

where A_α^* and $h\nu$ being the Tauc constant and incoming photon energy in the Tauc's law, respectively. $\Omega = 1/2$ for indirect transition and $\Omega = 2$ for direct transition¹¹. The necessary parameters $A_\alpha^* = 2.17 \times 10^5$ ($A_\alpha^* = 2.57 \times 10^4$) for SnS (ZIS) are obtained from refs¹² and¹³. Therefore, the total light absorptance in SnS/ZIS heterostructures can be obtained as

$$A_{abs} = A_{abs}^{SnS} + A_{abs}^{ZIS} \quad (10)$$

S2. Characterizations of crystal structure and element valance states the pristine SnS produced by PLD

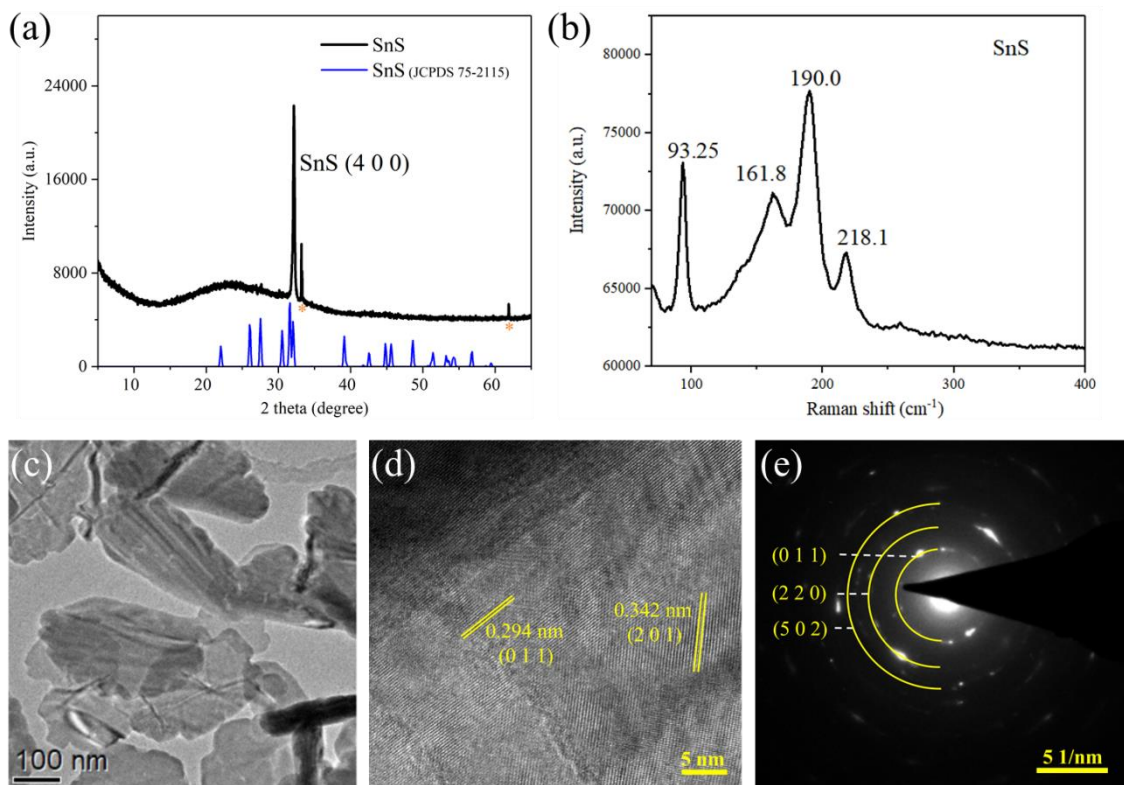


Fig. S1 Characterizations of the PLD-derived SnS. (a) 2θ - ω XRD pattern. The stars mark the diffraction peaks from the Si substrate. (b) Raman spectrum under 514 nm laser excitation. (c) Low-magnification TEM image. (d) High-resolution TEM image. (e) SAED pattern.

S3. Component characterizations of the pristine SnS produced by PLD

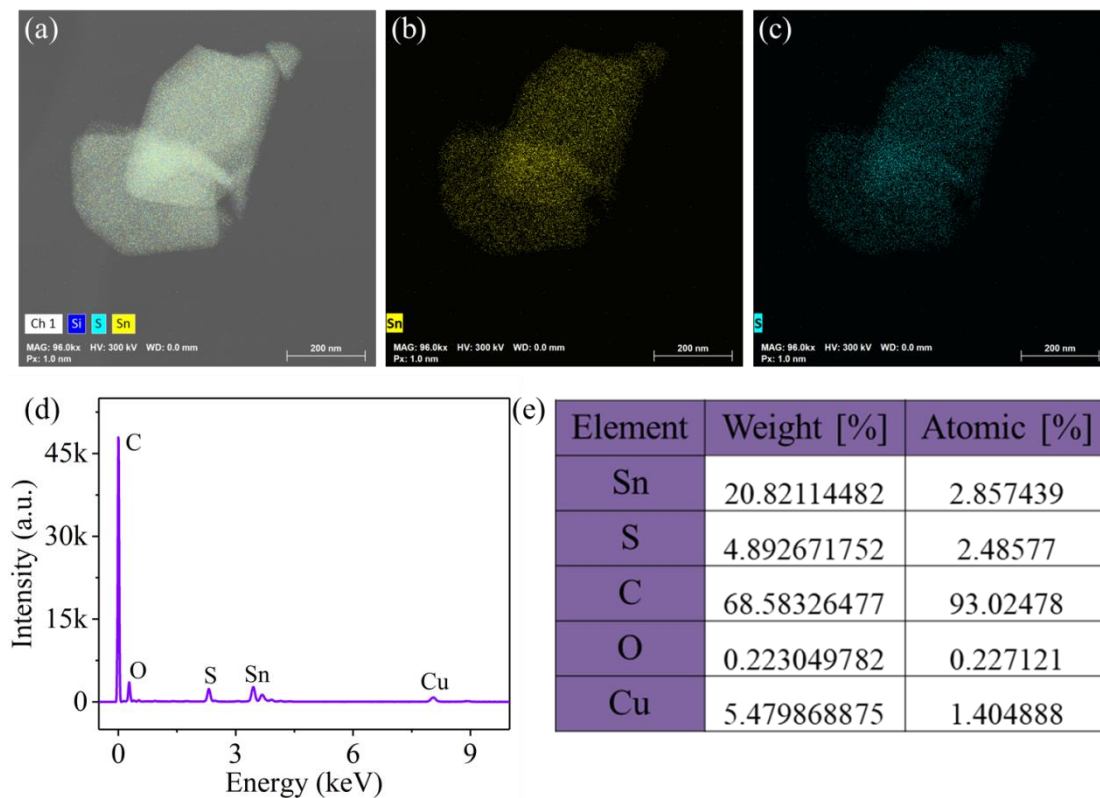


Fig. S2 (a) Low-magnification TEM image and the corresponding element mapping images of (b) Sn and (c) S elements. (d) EDS spectrum and (e) a summary of the corresponding element compositions.

S4. Fabrication process for preparing the SnS/ZIS hierarchical heterostructure

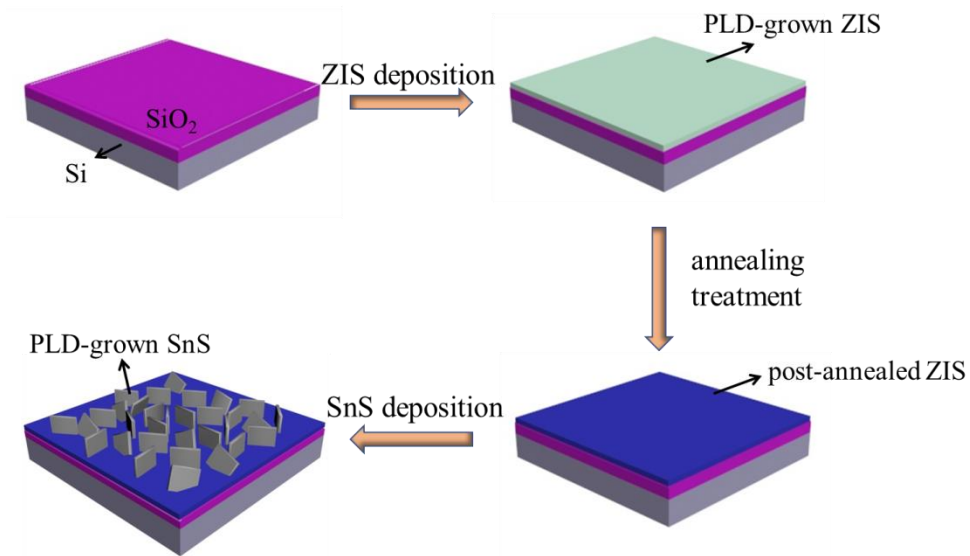


Fig. S3 Schematic illustration of the fabrication procedures for constructing SnS/ZIS *via* PLD.

S5. SEM morphology of the PLD-derived SnS/ZIS hierarchical heterostructure

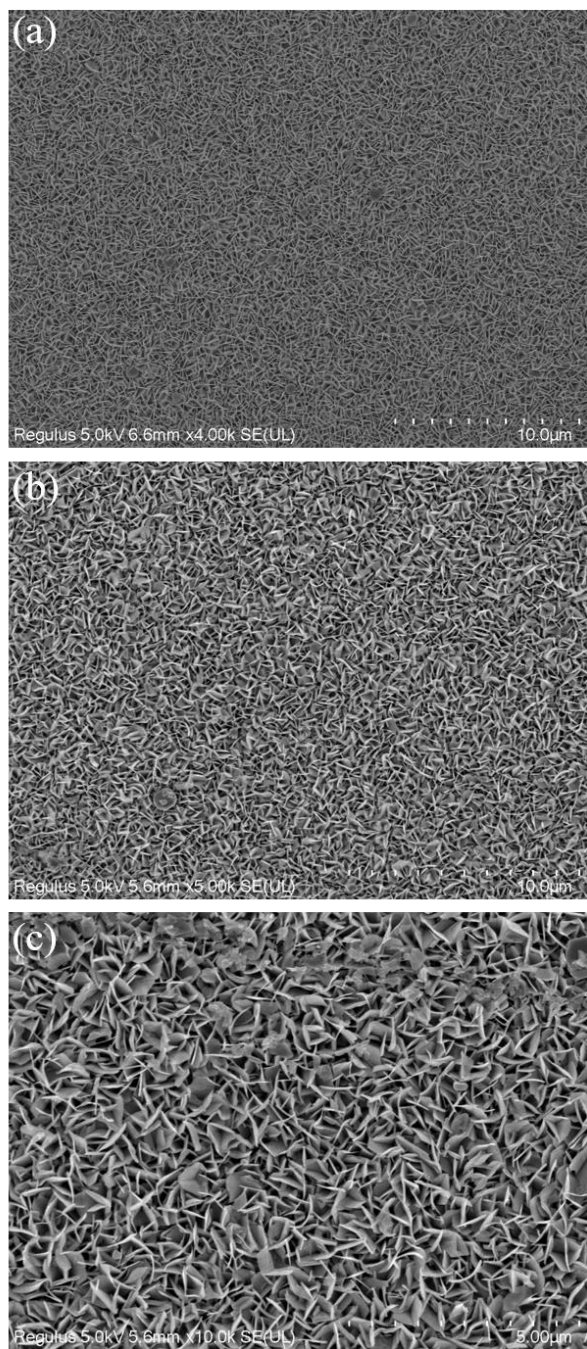


Fig. S4 SEM images of the PLD-derived SnS/ZIS hierarchical heterostructure. (a) 4000 \times , (b) 5000 \times , (c) 10000 \times .

S6. Crystalline orientation of the PLD-derived SnS on a SiO₂/Si substrate

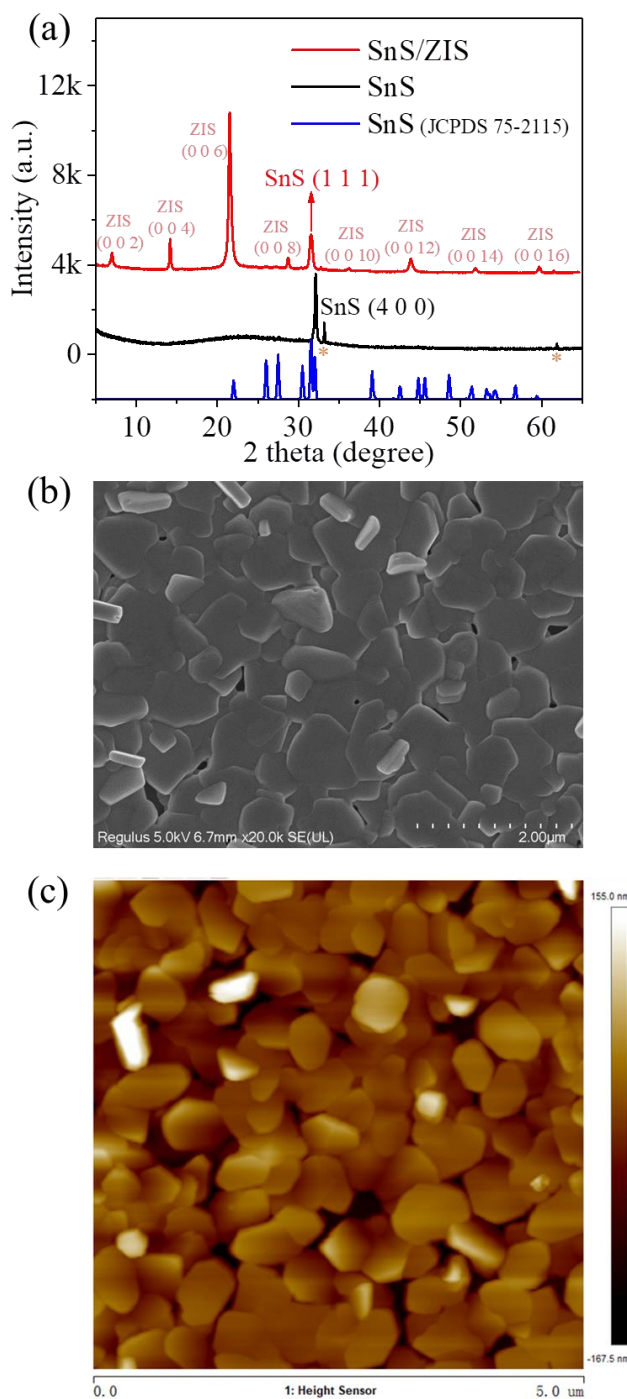


Fig. S5 (a) XRD patterns of the PLD-grown SnS (black line) and SnS/ZIS (red line) nanofilms. (b) SEM image (20000 \times) and (c) AFM image of the SnS grown on a SiO₂/Si substrate. The theoretical diffraction peaks of SnS are also included in (a) for comparison (blue line). Evidently, the SnS grown on a SiO₂/Si substrate is horizontally aligned, which is different from that grown on ZIS.

S7. Comparison of the surface roughness of the PLD-derived ZIS nanofilm and the SiO₂/Si substrate

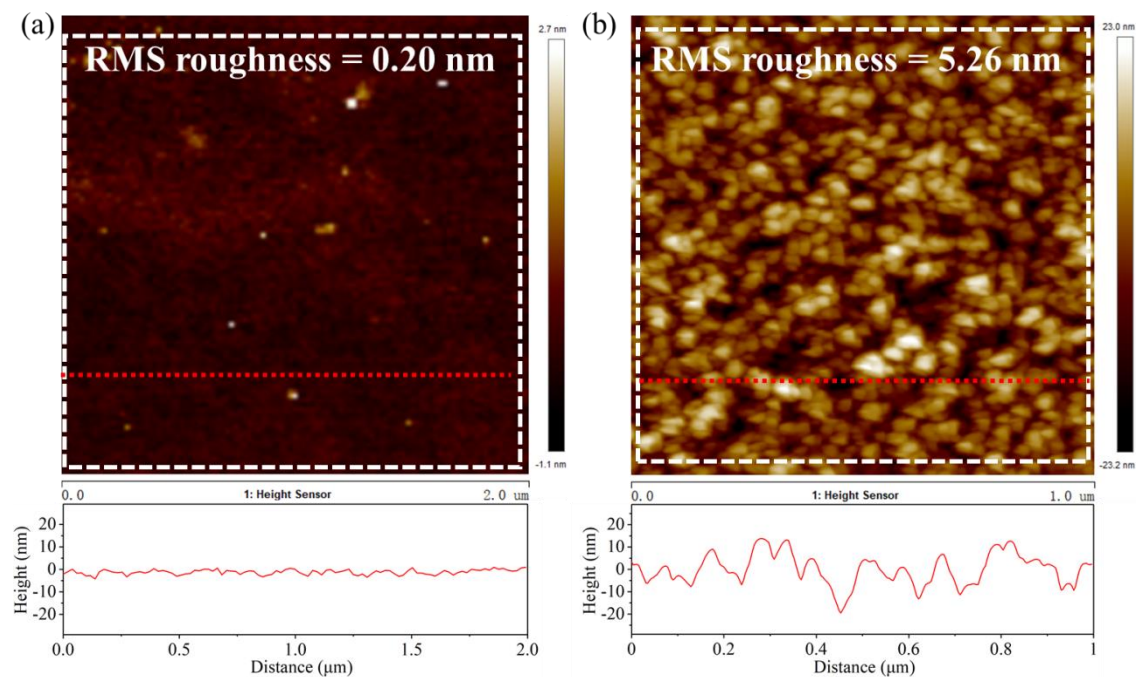


Fig. S6 AFM images of (a) a bare SiO₂/Si substrate and (b) a PLD-derived ZIS nanofilm. The bottom images present the corresponding line scans marked in the upper images. It is evident that the surface becomes much rougher after the deposition of ZIS nanofilm.

S8. Comparison of the reflectivity of the PLD-derived ZIS, SnS and SnS/ZIS nanofilms

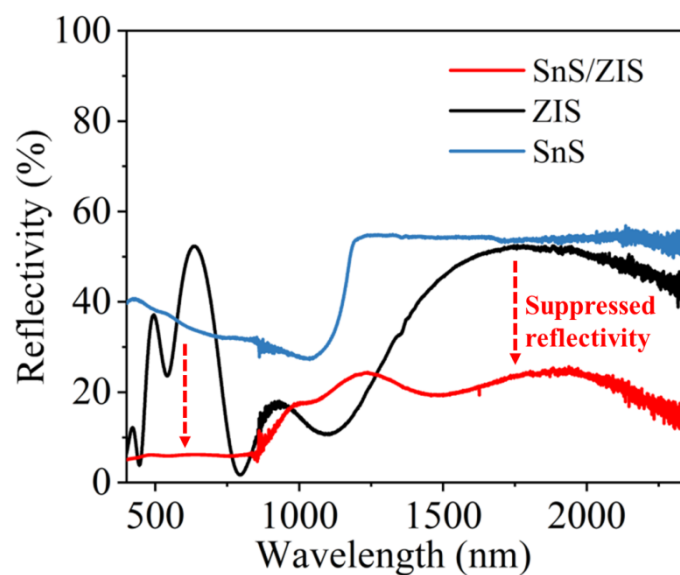


Fig. S7 Reflection spectra of the PLD-derived ZIS (black line), SnS (blue line) and SnS/ZIS (red line) nanofilms. The reflectivity of the SnS/ZIS nanofilm is largely suppressed (marked by the red arrows) as compared to the SnS and ZIS nanofilms.

S9. Morphology of the PLD-derived SnS/ZIS hierarchical heterostructure

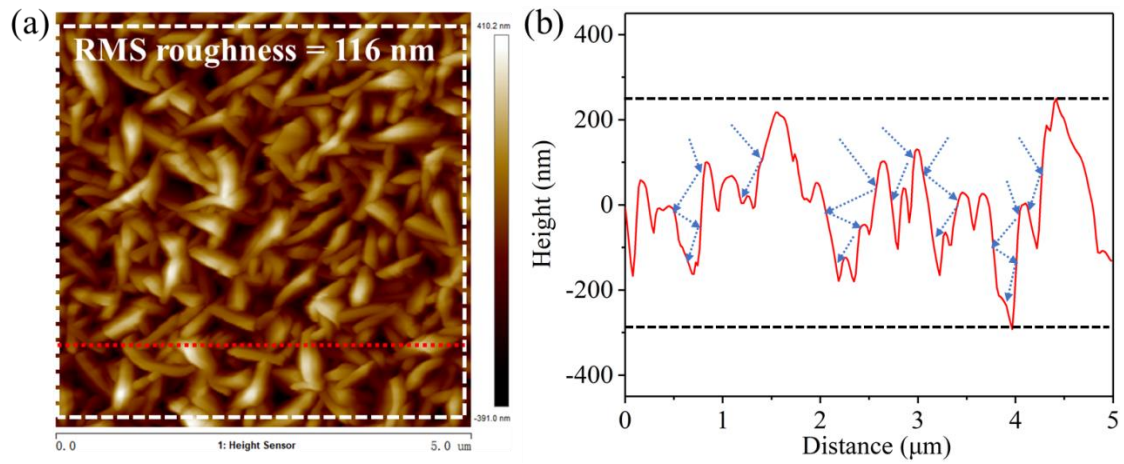


Fig. S8 (a) AFM image of the PLD-derived SnS/ZIS hierarchical heterostructure and (b) the corresponding height profile along the red dash line. The RMS roughness reaches up to ≈ 116 nm, which is ≈ 22 times of that of pristine ZIS. The maximum crest-to-valley amplitude exceeds 500 nm. The blue dash arrows indicate the light-trapping process.

S10. Schematic diagram illustrating the increase of reflectivity of SnS/ZIS at long wavelengths

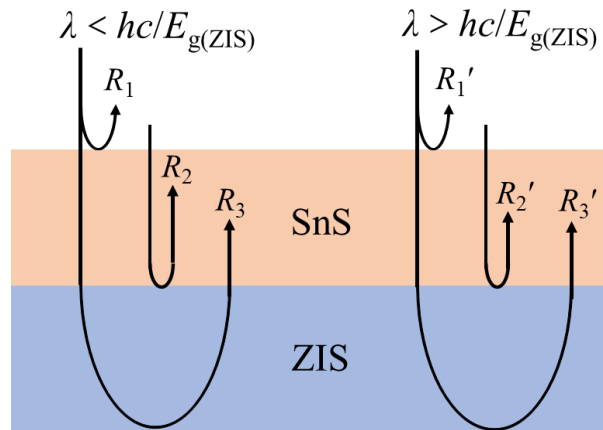


Fig. S9 Schematic diagram illustrating the increase of reflectivity of the PLD-derived SnS/ZIS hierarchical nanofilm at long wavelengths. Herein, R_1 , R_2 and R_3 represent the reflected light with wavelength below $hc/E_{g(\text{ZIS})}$ (h is the Planck constant, c is the light velocity, and $E_{g(\text{ZIS})}$ is the bandgap of ZIS) at the air/SnS, SnS/ZIS and ZIS/substrate interfaces, while R_1' , R_2' and R_3' represent the reflected light with wavelength beyond $hc/E_{g(\text{ZIS})}$ at the air/SnS, SnS/ZIS and ZIS/substrate interfaces.

S11. Power density dependent photocurrent of the ZIS and SnS/ZIS photodetectors upon various illuminations

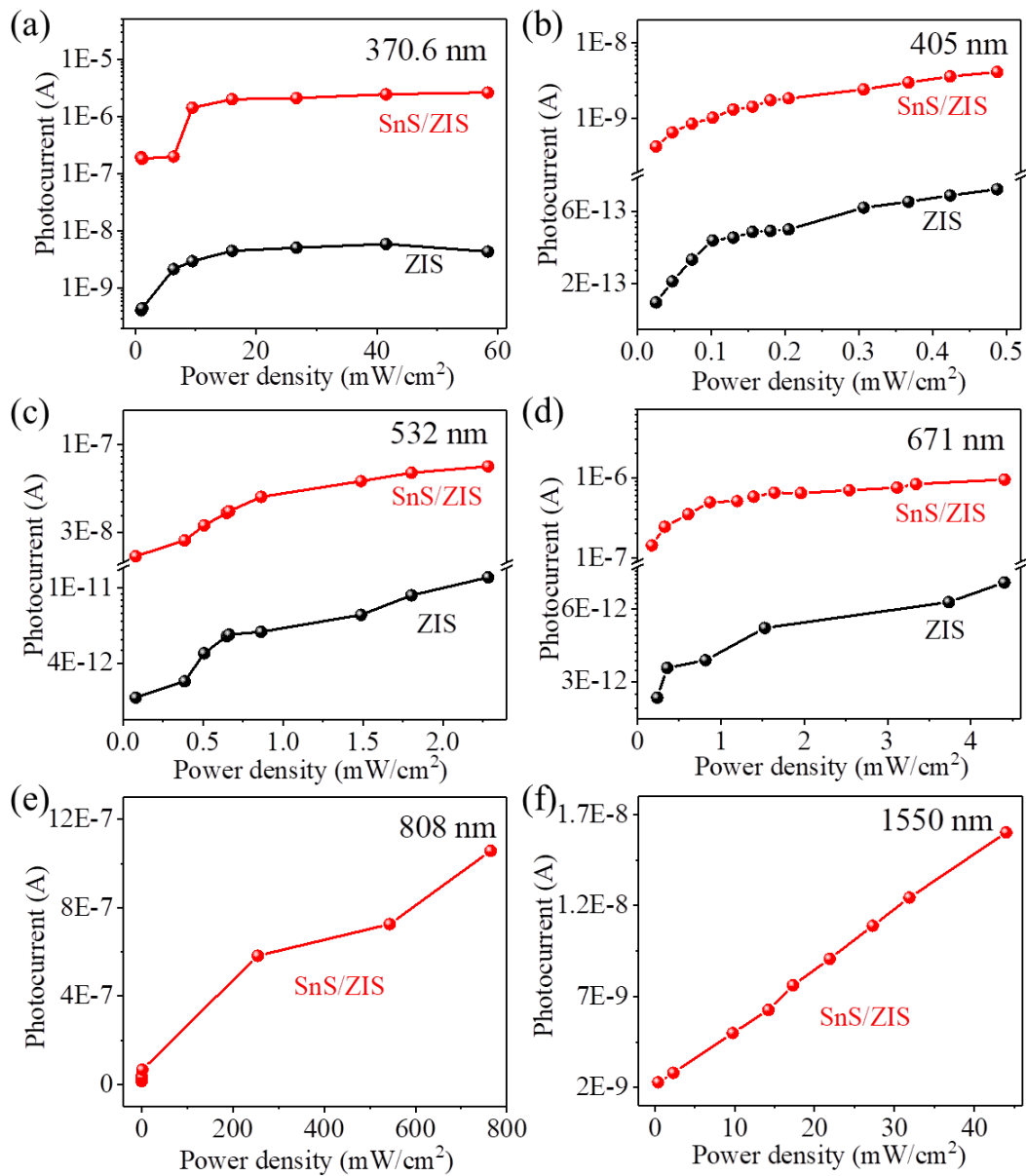


Fig. S10 Photocurrent as a function of the light power density of the ZIS and SnS/ZIS photodetectors upon (a) 370.6 nm, (b) 405 nm, (c) 532 nm, (d) 671 nm, (e) 808 nm and (f) 1550 nm illuminations. Upon long-wavelength illuminations (808 and 1550 nm), the ZIS photodetector exhibits no photoresponse.

S12. Definition of responsivity, EQE, detectivity, and rise/decay time

To evaluate the photosensitivity of photodetectors, several pivotal figures-of-merit have been developed, including responsivity (R), EQE and detectivity (D^*). Responsivity is equal to the photocurrent generated upon light illumination with per unit power. It can be calculated according to the following formula

$$R = \frac{I_{\text{light}} - I_{\text{dark}}}{PS}, \quad (10)$$

where I_{light} is the channel current under light illumination, I_{dark} is the channel current in dark condition, P is the power density of incident light, and S is the photosensitive area of the photodetector. EQE refers to the ratio of the number of photocarriers crossing the photosensitive channel to the number of incident photons onto the photosensitive area. It can be calculated according to the following formula

$$\text{EQE} = \frac{I_{\text{light}} - I_{\text{dark}}}{e} / \left(\frac{PS}{h\nu} \right) = \frac{hcR}{\lambda e}, \quad (11)$$

where h is the Planck constant, ν is the light frequency, c is the light speed, λ is the wavelength of incident light, e is the elemental charge. Detectivity is a performance metric used to evaluate the capability of photodetectors to distinguish weak light signals. It is calculated according to the following formula

$$D^* = \frac{\sqrt{SR}}{\sqrt{2eI_{\text{dark}}}}. \quad (12)$$

Response rate is commonly evaluated by the rise time and decay time during photoswitching. They are defined as the time intervals required for the photocurrent to rise from 0% to 90% and to decay from 100% to 10%, respectively.

S13. Power density dependent EQE of the ZIS and SnS/ZIS photodetectors upon various illuminations

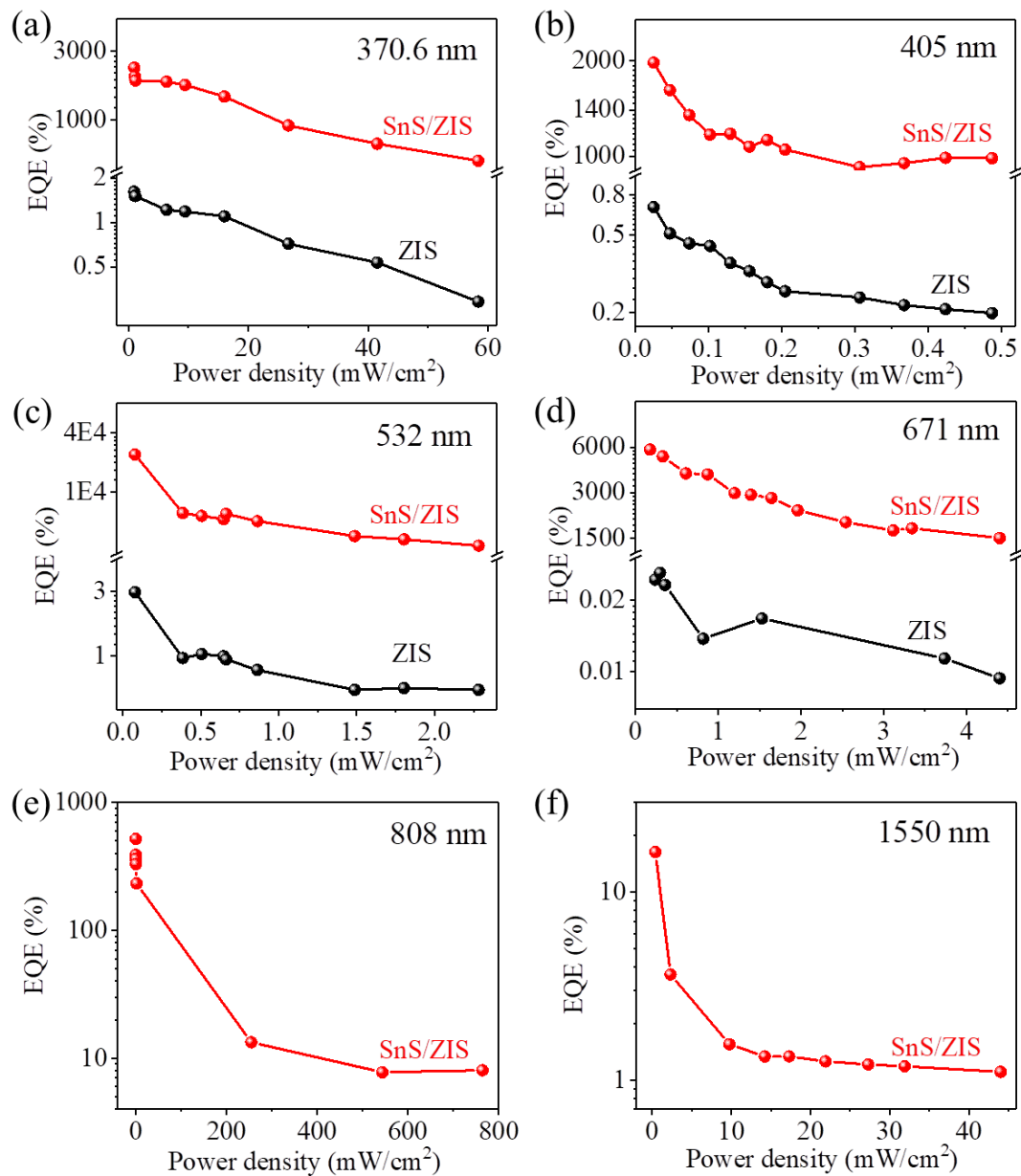


Fig. S11 EQE as a function of the power density of the ZIS and SnS/ZIS photodetectors upon (a) 370.6, (b) 405, (c) 532, (d) 671, (e) 808 and (f) 1550 nm illuminations. Upon long-wavelength illuminations (808 and 1550 nm), the ZIS photodetector exhibits no photoresponse.

S14. Power density dependent detectivity of the ZIS and SnS/ZIS photodetectors upon various illuminations

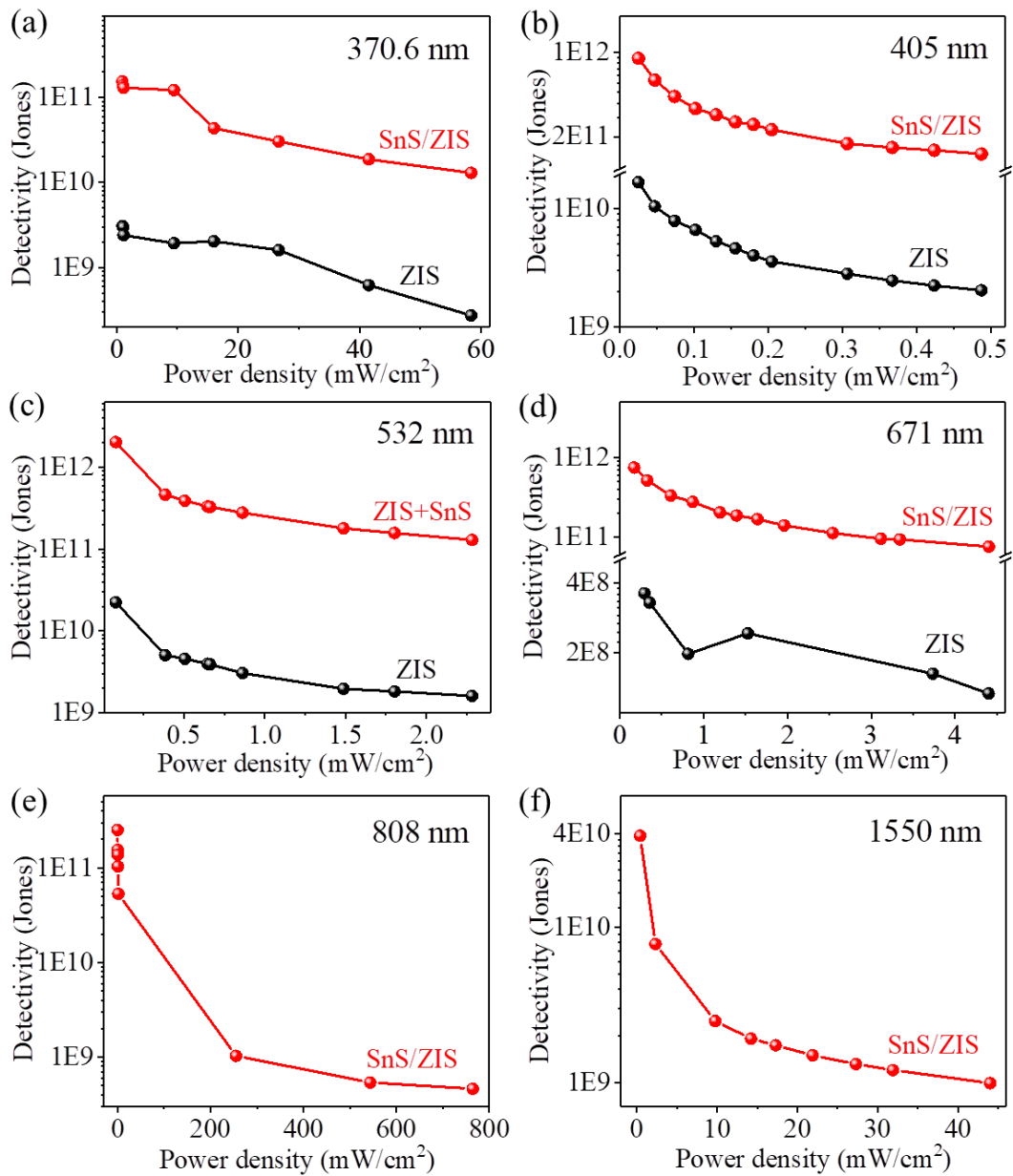


Fig. S12 Detectivity as a function of the power density of the ZIS and SnS/ZIS photodetectors upon (a) 370.6, (b) 405, (c) 532, (d) 671, (e) 808 and (f) 1550 nm illuminations. Upon long-wavelength illuminations (808 and 1550 nm), the ZIS photodetector exhibits no photoresponse.

S15. Photoswitching curves of the ZIS and SnS/ZIS photodetectors upon infrared illuminations

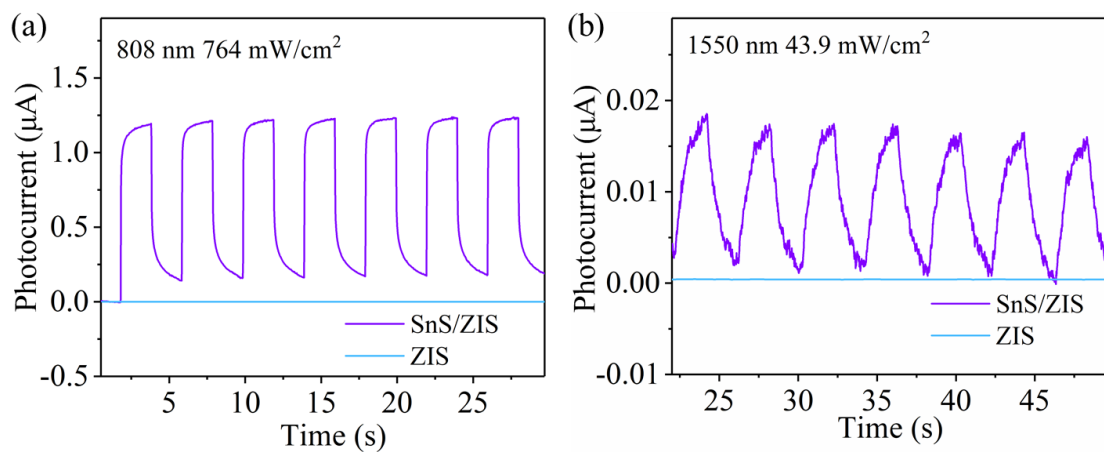


Fig. S13 Photoswitching curves of the pristine ZIS photodetector (wathet blue lines) and the SnS/ZIS photodetector (purple lines) upon (a) 808 and (b) 1550 nm periodic illuminations.

S16. Comparison of the spectral photoresponse of the ZIS and SnS/ZIS photodetectors

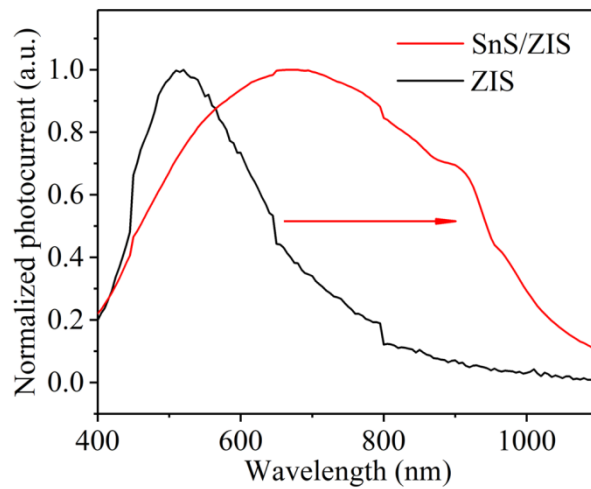


Fig. S14 Spectral photoresponse curves of the PLD-derived ZIS (black) and SnS/ZIS (red) photodetectors. It is evident that the effective optical window has been substantially extended to the long-wavelength region (marked by the red arrow) after the integration of SnS onto ZIS.

S17. Comparison of the enhancement factors of this study with state-of-the-art improvement strategies on 2DLM photodetectors

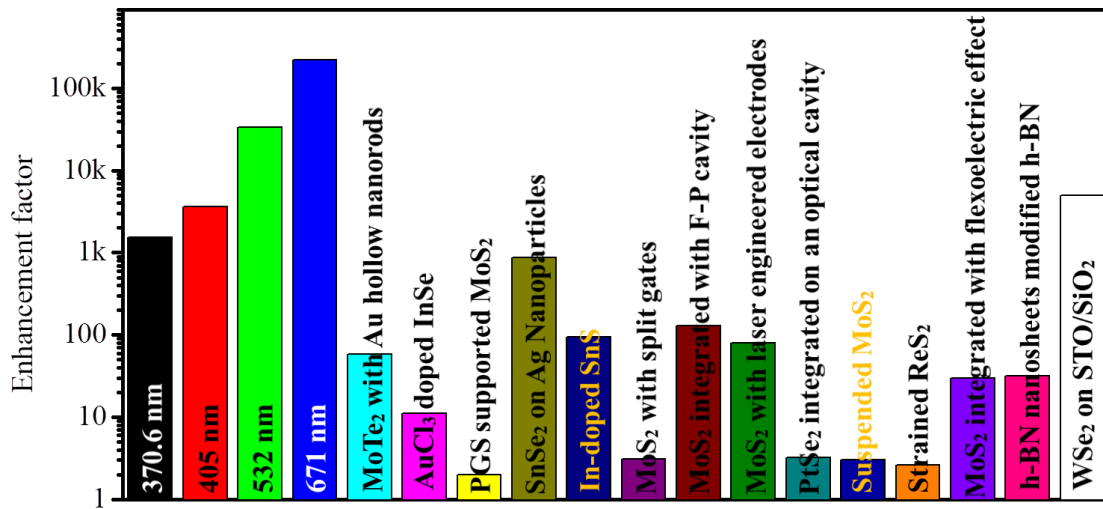


Fig. S15 A summary of the enhancement factors of our SnS/ZIS photodetector and the state-of-the-art improvement strategies on 2D material photodetectors.¹⁴⁻²⁷

S18. Response rate of a SnS/ZIS photodetector

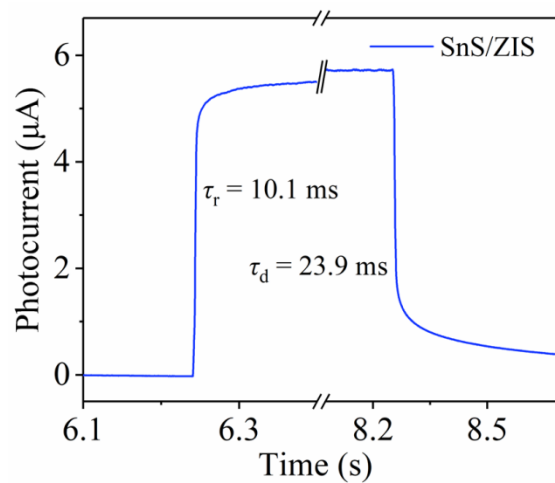


Fig. S16 Rise and fall edges of a SnS/ZIS photodetector.

S19. Comparison of the figures-of-merit of the SnS/ZIS photodetectors with state-of-the-art 2DLM based photodetectors

Table S1. A summary of the operating voltage (V_{ds}), responsivity, EQE, detectivity, effective wavelength range and response/recovery time of the SnS/ZIS photodetectors and state-of-the-art 2DLM based photodetectors.

Materials	V_{ds} (V)	R (A/W)	EQE (%)	D^* (Jones)	τ_r/τ_d (ms)	Wavelength (nm)	Ref.
SnS/ZIS	2	103.7	2.4×10^4	2.03×10^{12}	10.1/23.9	370–1550	Ours
WS ₂	10	0.144	N/A ^{a)}	10^8	0.07	400–800	28
TaIrTe ₄	N/A	0.13	4.03	N/A	N/A	633–4000	29
GaS	20	4.7	1312	1.4×10^{12}	<66	275–532	30
CuInP ₂ S ₆	4	0.0108	4.78	7.38×10^{10}	0.36/0.44	280–520	31
InSiTe ₃	11	0.0709	N/A	7.59×10^9	$\approx 5 \times 10^4$	365–1310	32
Si ₂ Te ₃	1	65	1.99×10^4	2.81×10^{12}	210/478	405–1064	33
SnSe ₂ /AgNPs ^{b)}	0.1	0.194	59.4	8.4×10^9	21	405–2200	17
SiAs	2	0.016	1200	1×10^{10}	3000/1500	325–514.5	34
SiP	N/A	≈ 18	6115	$\approx 2.25 \times 10^{11}$	0.06/0.03	300–700	35
In _{4/3} P ₂ Se ₆	5	4.31	1485.2	6.3×10^{12}	0.47/0.44	360–639	36
Au ₂ P ₃	N/A	0.97	77.6	2.9×10^{10}	1440/1340	1550–2611	37
MoTe ₂	–2	0.5	37	N/A	N/A	1550	38
WSe ₂	2	170	3962	1.1×10^{12}	0.014	432	19
PtSe ₂	0.05	0.054	8.02	2.5×10^6	5.4×10^{-5}	532–8350	22
MoS ₂ /PSS ^{c)}	20	8.8×10^{-3}	3	1.2×10^{10}	2240/2270	365–660	23
h-BN/AuNPs ^{d)}	35	2.24×10^{-5}	0.14	N/A	N/A	205–390	26
MoS ₂ /Au Array	5	13	N/A	N/A	N/A	532–1550	39

a) N/A: Not applicable. b) AgNPs: Ag nanoparticles.

c) PSS: Patterned sapphire substrate. d) AuNPs: Au nanoparticles.

S20. Comparison of the figures-of-merit of the SnS/ZIS photodetectors with state-of-the-art 2DLM based heterojunction photodetectors

Table S2. A summary of the operating voltage (V_{ds}), responsivity, EQE, detectivity, effective wavelength range and response/recovery time of the SnS/ZIS photodetectors and state-of-the-art 2DLMs based heterojunction photodetectors.

Heterostructures	V_{ds} (V)	R (A/W)	EQE (%)	D^* (Jones)	τ_r/τ_d (ms)	Wavelength (nm)	Ref.
SnS/ZIS	2	103.7	2.4×10^4	2.03×10^{12}	10.1/23.9	370–1550	Ours
Sn/InSe	0	0.032	8.7	1.7×10^{11}	30/37	300–700	40
MoS ₂ QDs ^{a)} /MoS ₂	2	0.0128	2.86	7.2×10^9	2740	UV-vis-NIR	41
PtSe ₂ /Gr ^{b)}	0.02	0.4	76.5	$\approx 10^7$	50.6/37.3	637–10600	42
Te/MoS ₂	0.8	28.4	5700	2.7×10^{10}	N/A ^{c)}	980–3000	43
PtS ₂ /WSe ₂	1	1.7	≈ 3320	3.8×10^{10}	0.008/0.009	400–800	44
Te/ReS ₂	1	180	≈ 35316	7.2×10^9	5/8	632	45
WS ₂ /GaS _{0.87}	2	25	≈ 4984	1.5×10^9	5000	395–622	46
WO ₃ /WSe ₂	2	3663	N/A	N/A	50	White light	47
WSe ₂ /WS ₂	N/A	300	6.99×10^4	4.3×10^{10}	1.46/1.42	532	48
MoTe ₂ /Gr	–0.1	0.2	19.08	N/A	19×10^{-9}	1265–1330	49
CuInSe ₂ /In ₂ Se ₃	0	0.01052	3.22	N/A	195/180	405–1310	50
Bi ₂ Se ₃ /BP ^{d)}	0.1	0.0121	≈ 1.58	N/A	1.3/8.4	350–950	51
MoS ₂ /Si	6	0.01007	≈ 1.46	4.53×10^{10}	0.078/0.076	850	52
Bi ₂ O ₂ Se/CsPbBr ₃	3	46	≈ 9200	2.73×10^{10}	0.012/0.028	390–840	53
Te/Bi ₂ O ₂ Se	0.1	130	≈ 30300	2.5×10^{11}	0.33/0.43	405–635	54
InSe/NbTe ₂	2	84	≈ 19579	N/A	< 10	532	55
Cs ₂ AgBiBr ₆ /WS ₂ /Gr	4	0.054	≈ 14.7	1.13×10^{12}	0.0523/0.0536	360–660	56

a) QDs: quantum dots. b) Gr: graphene. c) N/A: Not applicable.

d) BP: black phosphorus.

S21. Bandgap values of ZIS and SnS deduced from the absorption spectra

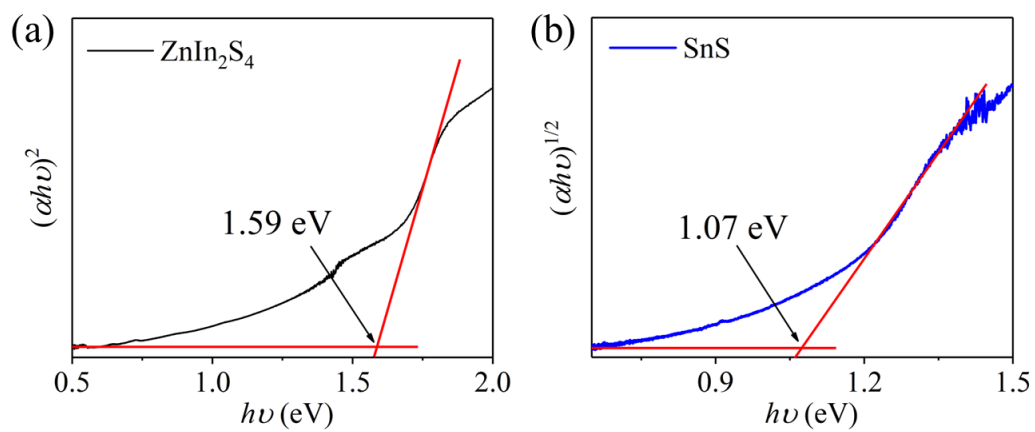


Fig. S17 Tauc plots of the absorption spectra of the PLD-derived (a) ZIS and (b) SnS nanofilms.

S22. Band alignment of the PLD-derived ZIS and SnS

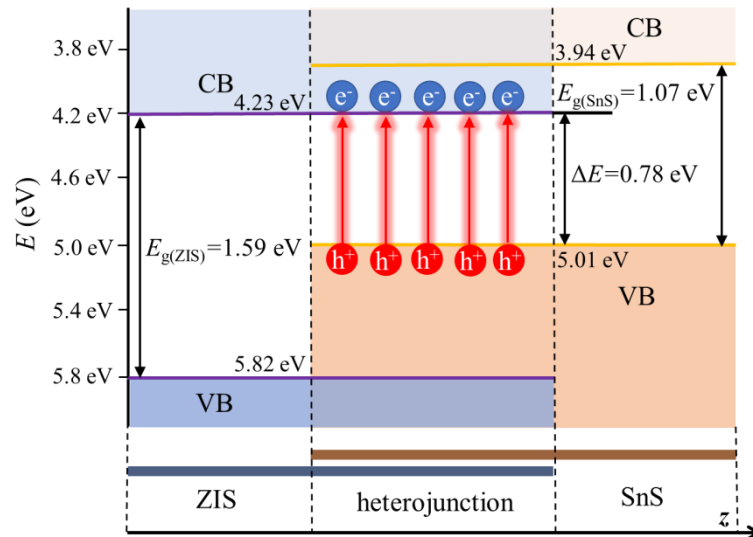


Fig. S18 Band alignment of the PLD-derived ZIS and SnS deduced from the UPS and absorption spectra results. The red arrows represent interlayer excitation. Red balls: holes. Blue balls: electrons. CB: conduction band. VB: valence band. $E_{g(\text{ZIS})}$: bandgap of ZIS. $E_{g(\text{SnS})}$: bandgap of SnS. ΔE : bandgap of SnS/ZIS heterojunction.

S23. Photoresponse of the SnS/ZIS photodetector to 2240 nm illumination

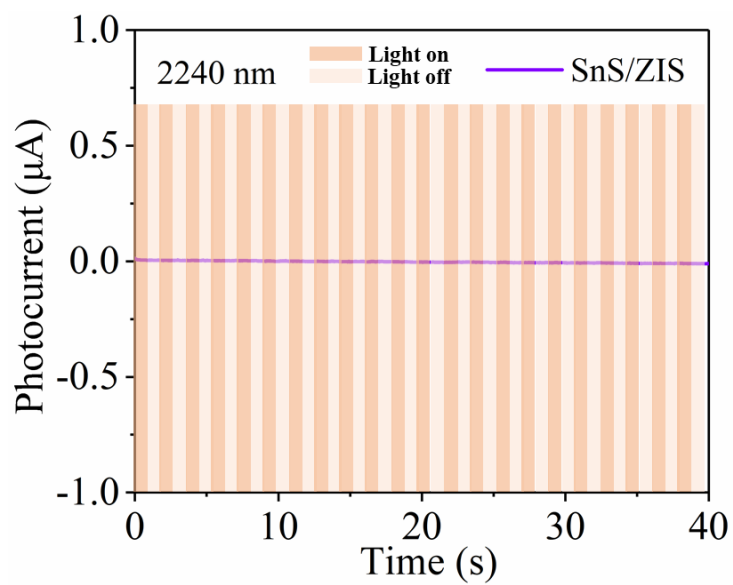


Fig. S19 Photocurrent as a function of time of the SnS/ZIS photodetector under periodic 2240 nm illumination. No discernable photoresponse is observed.

S24. The effect of the laser pulse number of SnS on the density of the SnS nanosheets

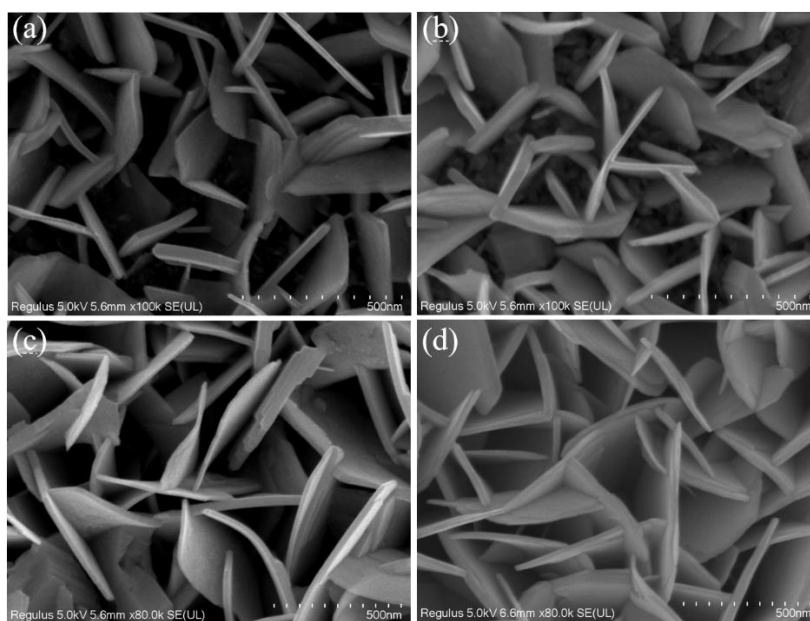


Fig. S20 SEM images of the PLD-derived SnS/ZIS hierarchical heterostructure with the pulse number of SnS of (a) 1000, (b) 2000, (c) 3000, and (d) 4000, respectively.

S25. The effect of the pulse number of SnS on the photoresponse of the SnS/ZIS photodetectors

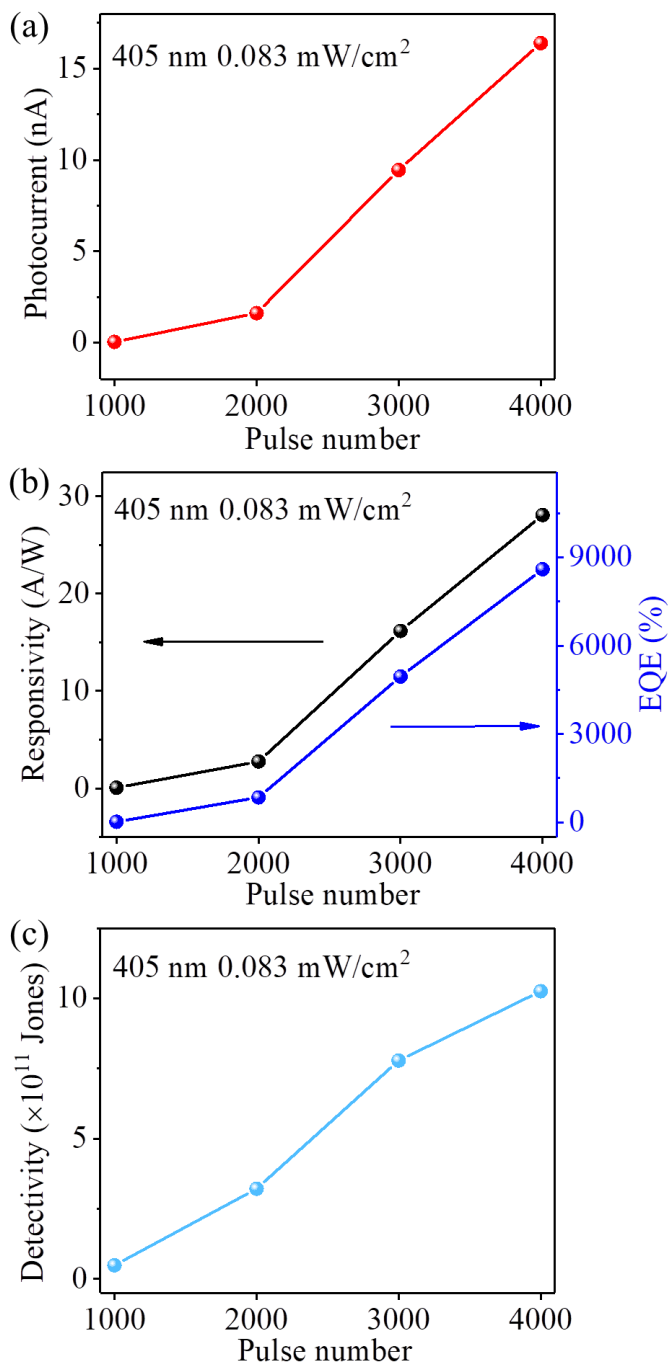


Fig. S21 (a) Photocurrent, (b) responsivity (black line) and EQE (blue line), and (c) detectivity as a function of the laser pulse number of the PLD-derived SnS/ZIS photodetectors.

S26. Theoretical band structures of ZIS and SnS

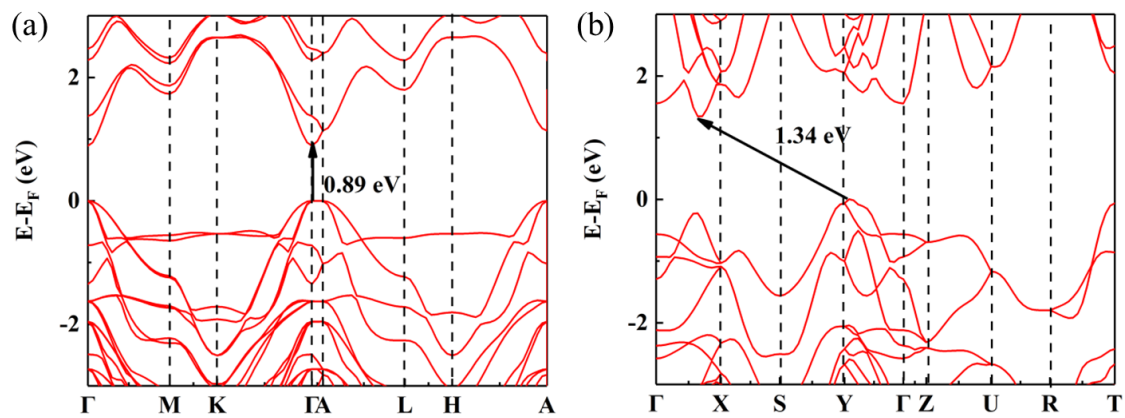


Fig. S22 Band structures of (a) bulk ZIS and (b) bulk SnS based on the framework of HSE06.

S27. Evaluation of the uniformity of the wafer-scale SnS/ZIS nanofilm

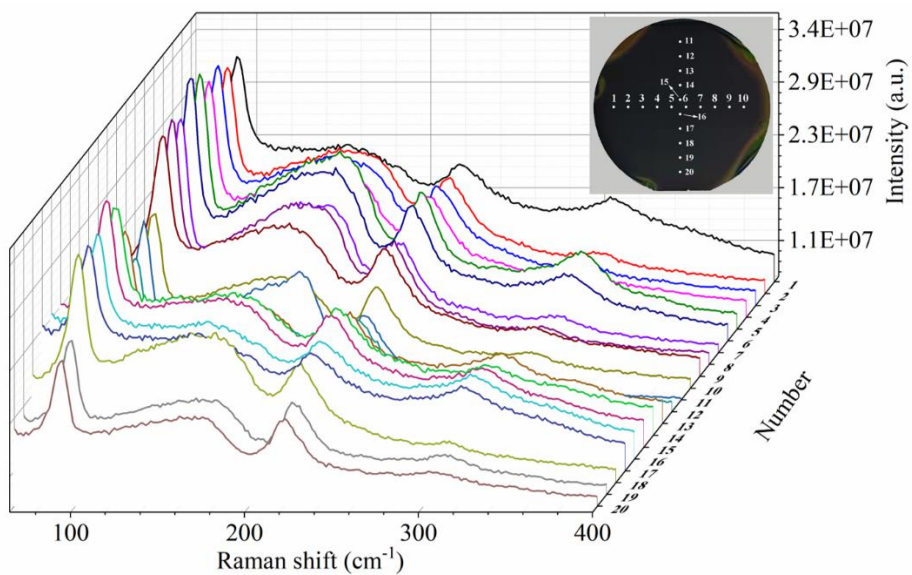


Fig. S23 A summary of the Raman spectra of random 20 points across the PLD-derived wafer-scale SnS/ZIS nanofilm upon 514 nm laser excitation. The inset presents the digital photograph marking the tested points.

S28. Optical imaging based on the SnS/ZIS photodetectors under various illuminations

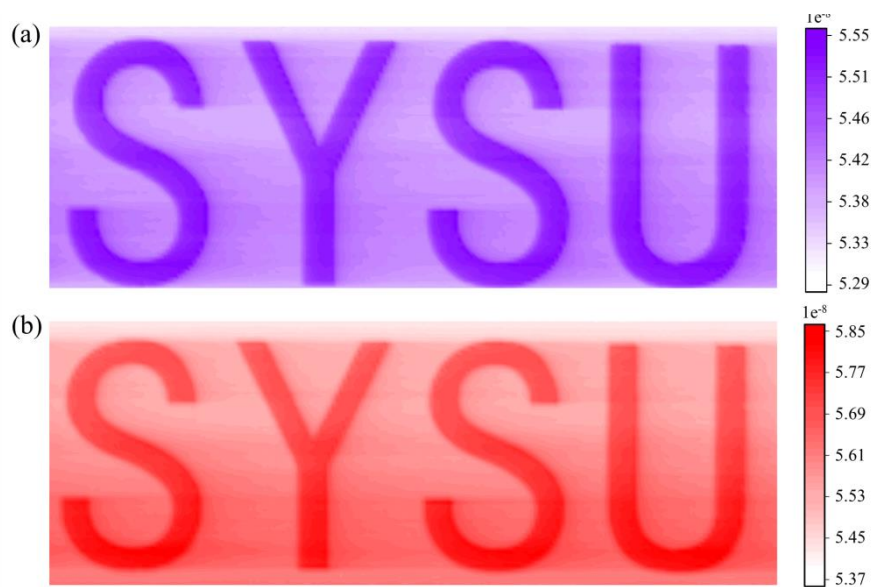


Fig. S24 Imaging results of the “SYSU” pattern by the SnS/ZIS photodetectors under (a) 405 (bluish violet) and (b) 635 nm (red) illuminations.

S29. Stability of the PLD-derived SnS/ZIS nanofilm

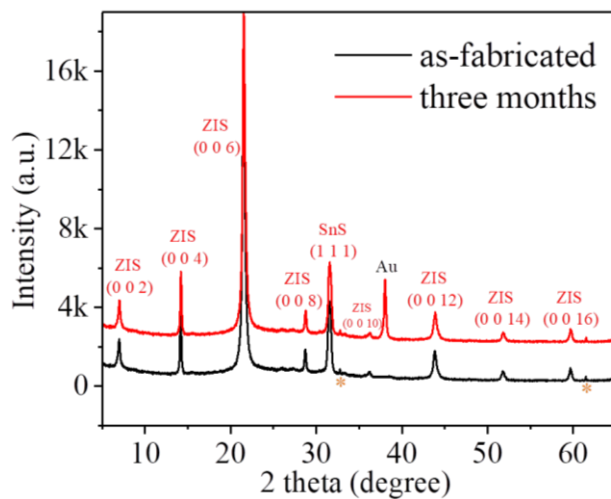


Fig. S25 XRD patterns of the as-fabricated SnS/ZIS nanofilm (black line) and that after storage in the ambient environment for 3 months (red line). All the diffraction peaks from both ZIS and SnS are well maintained.

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