Supplementary information

Narrow-bandgap Sn-Pb Mixed Perovskite Single Crystal for High-performance Near-Infrared Photodetector

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Table S1. Comparison of full-width at half-maximum of the $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal with previously reported perovskite single crystals.

Number	Materials	FWHM (°)	References
1	$Cs_2AgBiBr_6$	0.061	30
2	$MAPbBr_3$	0.042	31
3	$(PEA)_2PbI_4$	0.1055	32
4	$CsPbBr_3$	0.13	33
5	$MAPbBr_3$	0.013	34
6	$MAPbBr_3$	0.019	35
7	$MAPbI_3$	0.06	36
8	$MAPbCl_3$	0.0447	37
9	$MAPbBr_3$	0.0096	38
10	$MAPbCl_{0.7}Br_{2.3}$	0.016	39
11	$CsPbBr_3$	0.0042	40
12	$(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$	0.033	Our work

Table S2. Comparison of *LDR* of $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal NIR photodetector with previous Sn-Pb mixed perovskite devices.

Number	Materials	*LDR*	References
1	$(FASnI_3)_{0.6}(MAPbI_3)_{0.4}$	160	13
2	$MA_{0.975}Rb_{0.025}Sn_{0.65}Pb_{0.35}I_3$	110	15
3	$PEA_2MA_4(Sn_{0.5}Pb_{0.5})_5I_{16}$	158	17
4	$FA_{0.85}Cs_{0.15}Sn_{0.5}Pb_{0.5}I_3$	103	20
5	$(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$	163.5	Our work

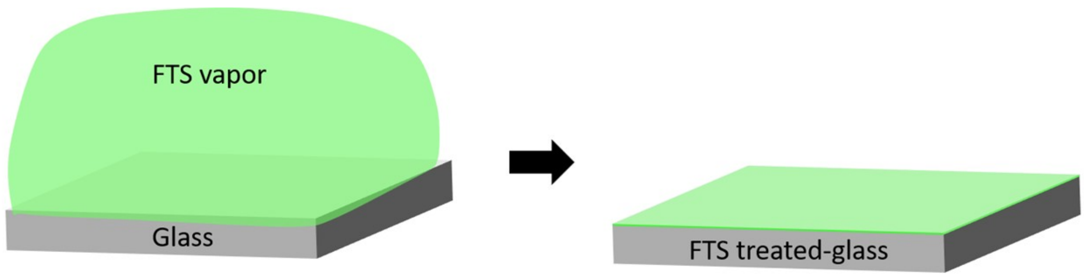

Figure S1. Schematic illustration of FTS treatment procedure. In this process, glass substrate was placed in the environment of FTS vapor to realize a hydrophobic surface with low surface energy.

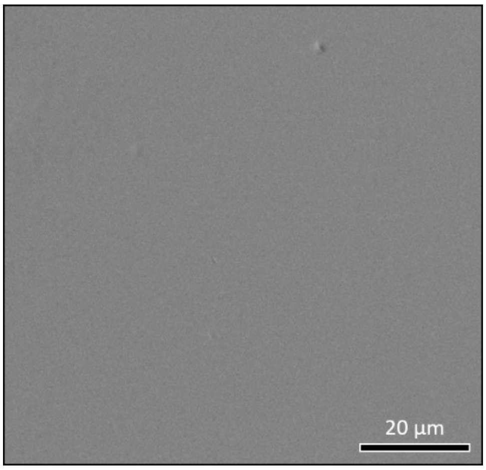

Figure S2. Zoom-in SEM image of the surface of the as-grown $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal.

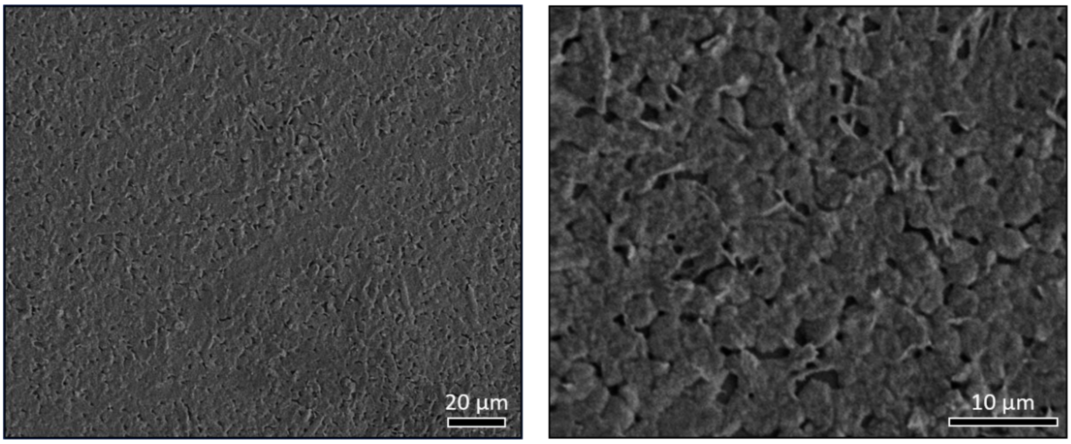

Figure S3. SEM images of the spin-coated $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ polycrystalline film, indicating abundant of grain boundaries and defects.

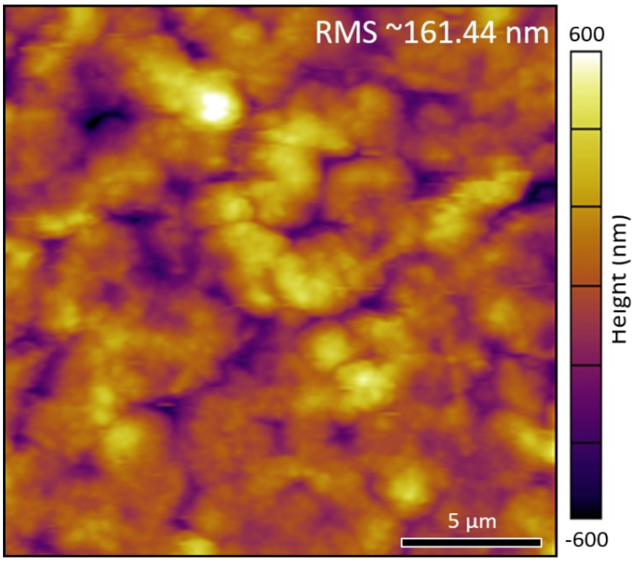

Figure S4. AFM image of the $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ polycrystalline film, which indicates a large RMS of 161.44 nm.

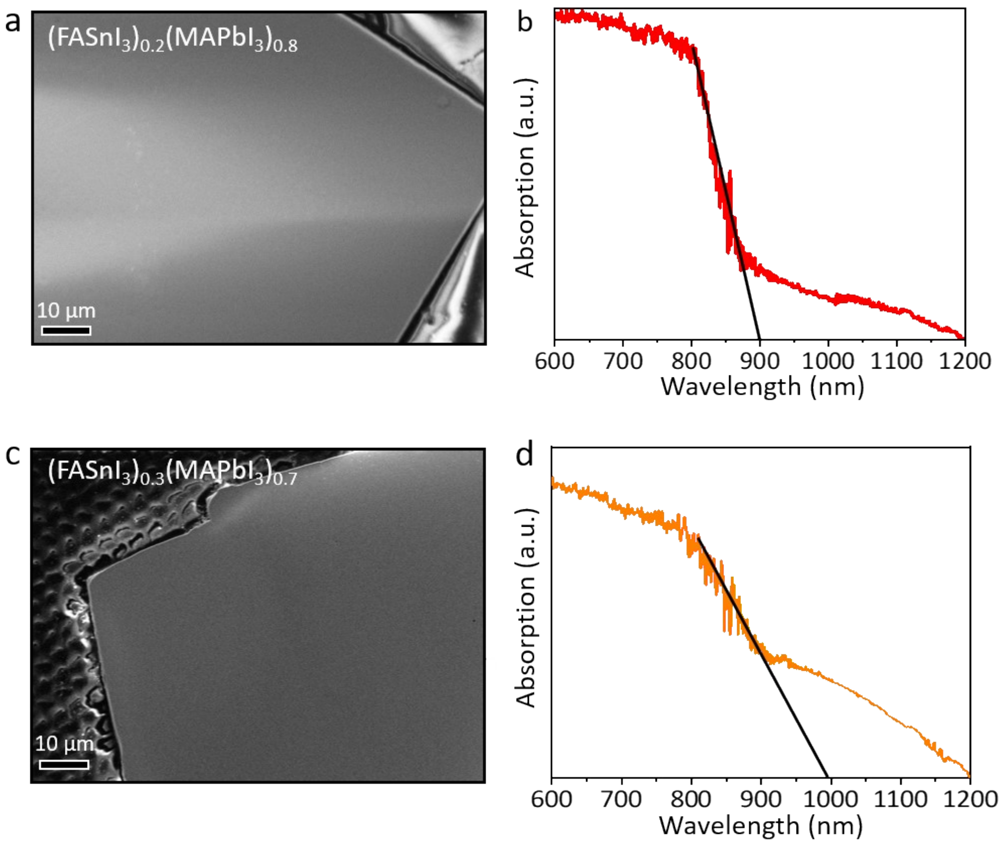

Figure S5. (a) SEM image and (b) corresponding absorption spectra of $(FASnI_3)_{0.2}(MAPbI_3)_{0.8}$ single crystal. (c) SEM image and (d) corresponding absorption spectra of $(FASnI_3)_{0.3}(MAPbI_3)_{0.7}$ single crystal.

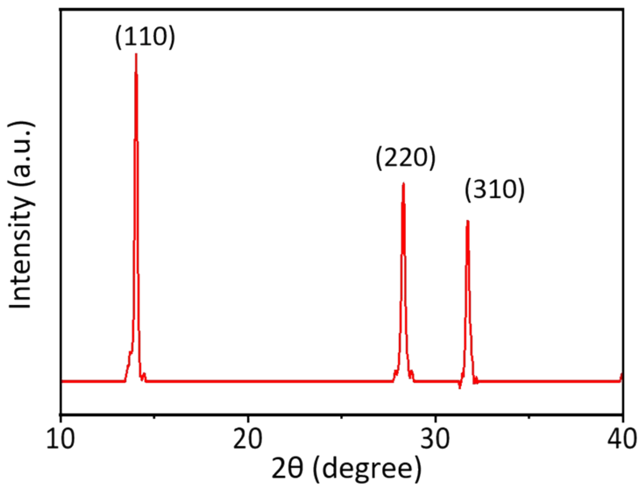

Figure S6. X-ray diffraction pattern of the $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ polycrystalline film, indicating multiple out-plane peaks.

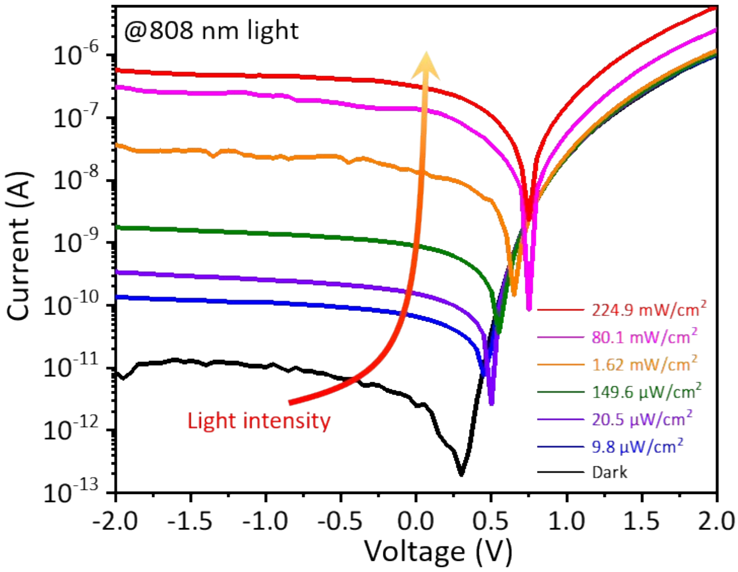

Figure S7. Current-voltage characteristics of the $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal NIR photodetector performed under dark condition and 808 nm light illumination by varying light intensities.

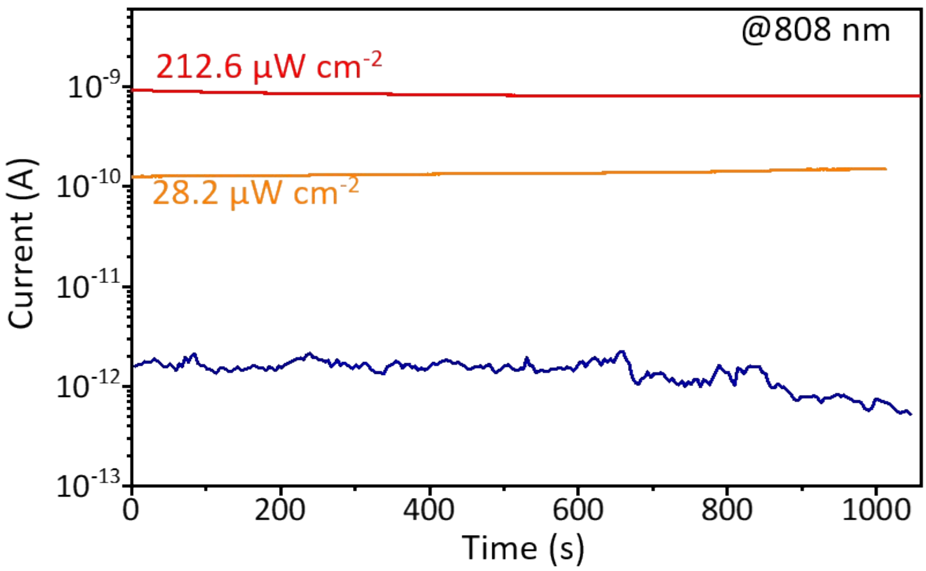

Figure S8. Long-term stability of the $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal NIR photodetector under light illumination.

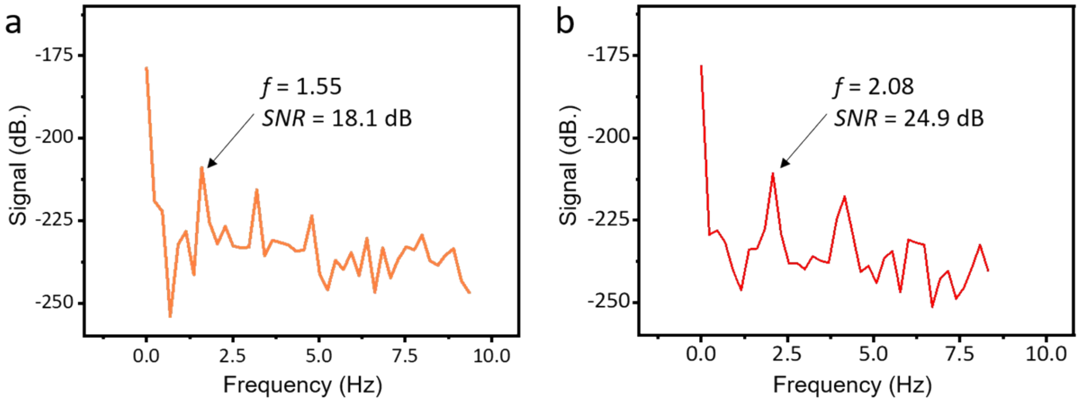

Figure S9. Fast Fourier transform of the PPG signals under at normal (a) and after-exercise (b) conditions.

Calculation of hole mobility and surface trap density

The $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ single crystal has excellent carrier mobility and low trap density. To verify this point, we carried out the space charge-limited current (SCLC) measurements. For SCLC method in lateral devices, a gap-type device was fabricated using single crystal with Au electrodes with channel width and length of 180 and 300 μm. From the *I*-*V* curve (Fig. 3c), we could obtain the carrier mobility for gap-type devices by the Geurst's SCLC model:[1]

$$I = \frac{2\mu\varepsilon_0\varepsilon_r W}{\pi}\left(\frac{V^2}{L^2}\right)$$

According to the Geurst theory the onset voltage (V_{TFL}) for a gap-type structure was equal to[2]

$$V_{TFL} = \frac{\pi\sigma_0 L}{4\varepsilon_0\varepsilon_r}$$

where μ is the carrier, σ_0 is the surface trap density per unit area, ε_0 is the vacuum permittivity and ε_r is relative dielectric constant of $(FASnI_3)_{0.1}(MAPbI_3)_{0.9}$ perovskite, 32.

1. Liu, Y. et al. Fast growth of thin $MAPbI_3$ crystal wafers on aqueous solution surface for efficient lateral-structure perovskite solar cells. *Adv. Funct. Mater.*, 2019, **29**, 1807707.
2. Zuleeg, R. & Knoll, P. Space-charge-limited currents in heteroepitaxial films of silicon grown on sapphire. *Appl Phys. Lett.*, 1967, **11**, 183-185.