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Supporting Information

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2D WSe_2 Flakes for Synergistic Modulation of Grain Growth and Charge Transfer in Tin-based Perovskite Solar Cells

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Figures:

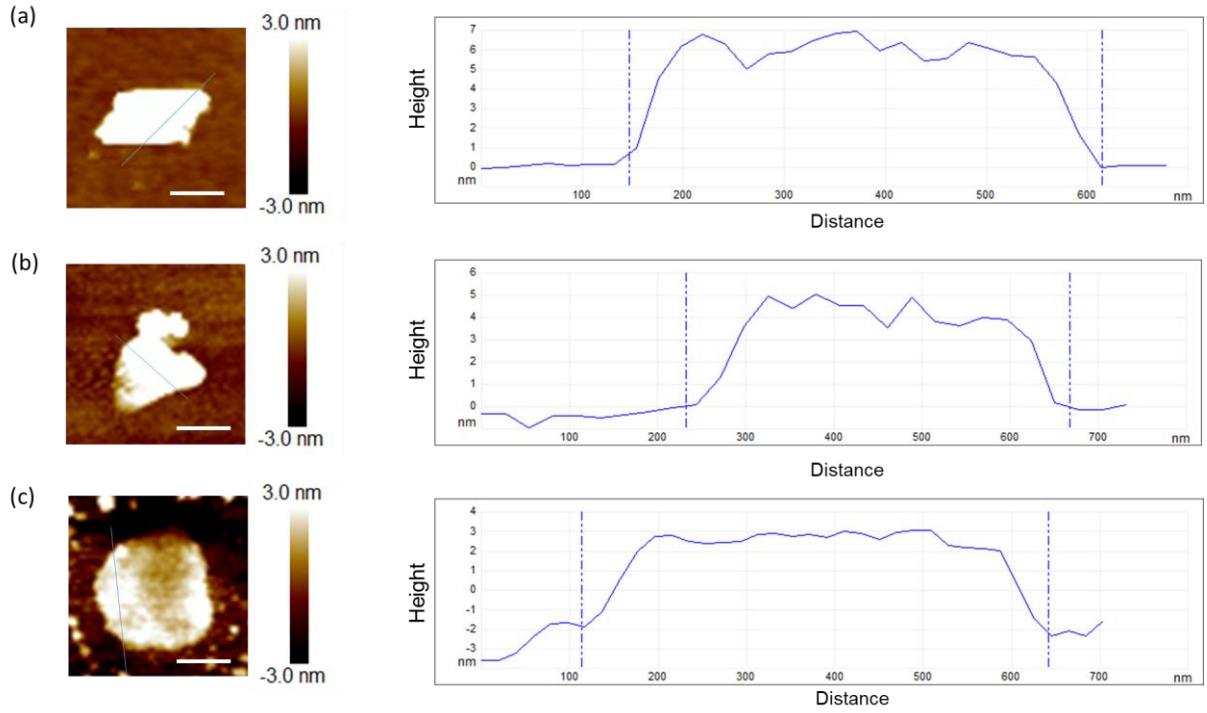

Figure S1 Representative AFM images of a) MoS_2, b) WS_2 and c) WSe_2 flakes deposited on Si substrates. Scale bar: 300 nm. The height profiles on the right were extracted from the indicated blue lines in the AFM images.

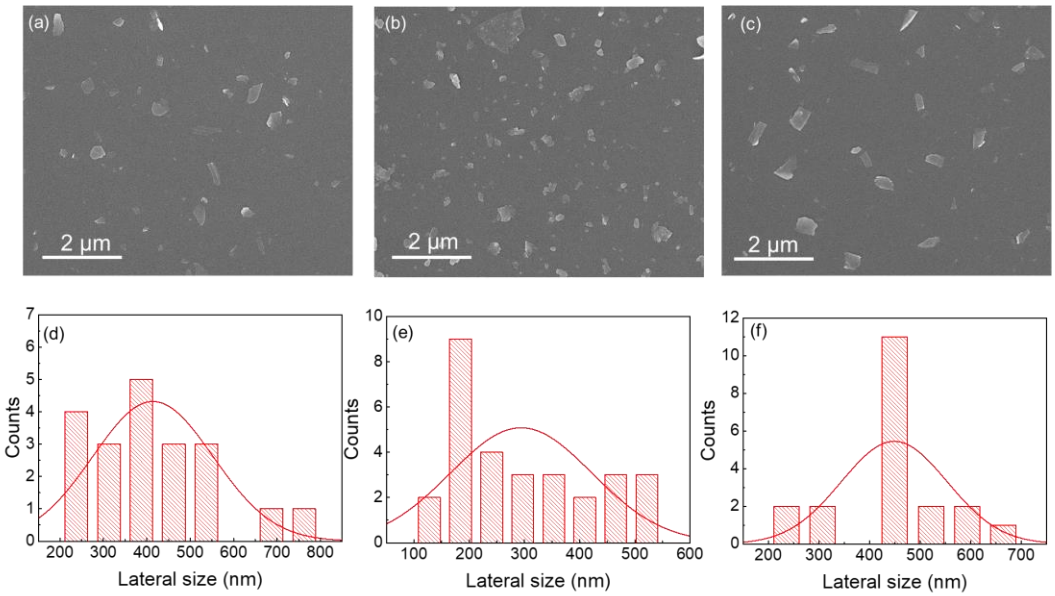

Figure S2 SEM images of as-exfoliated a) MoS_2, b) WS_2 and c) WSe_2 flakes spin-coated on Si-substrates. Lateral size distributions of d) MoS_2, e) WS_2 and f) WSe_2 flakes extracted from a-c).

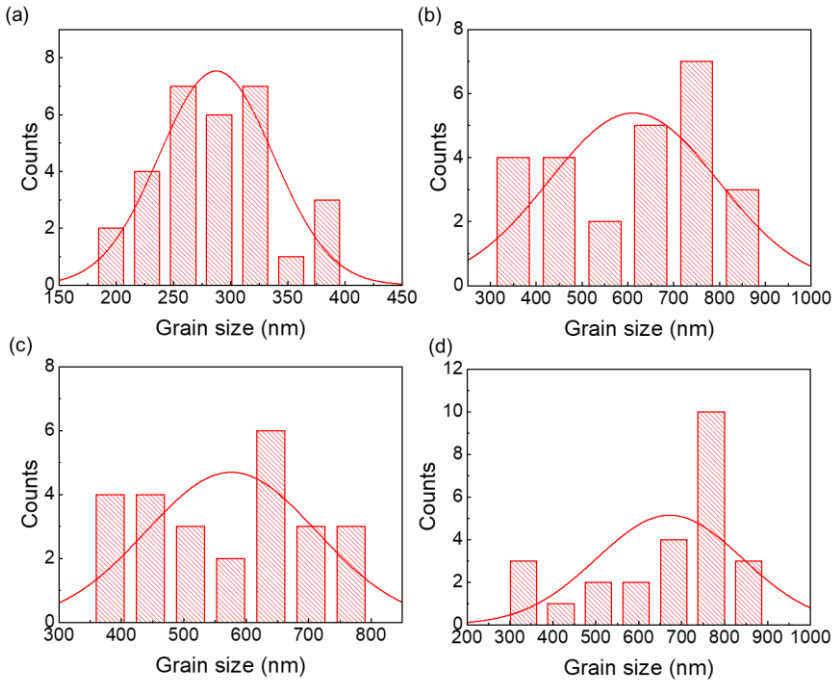

Figure S3 Statistical grain size distribution of $FASnI_3$ films deposited on a) NiO_x/ITO, as well as b) MoS_2, c) WS_2, and d) WSe_2 modified NiO_x/ITO.

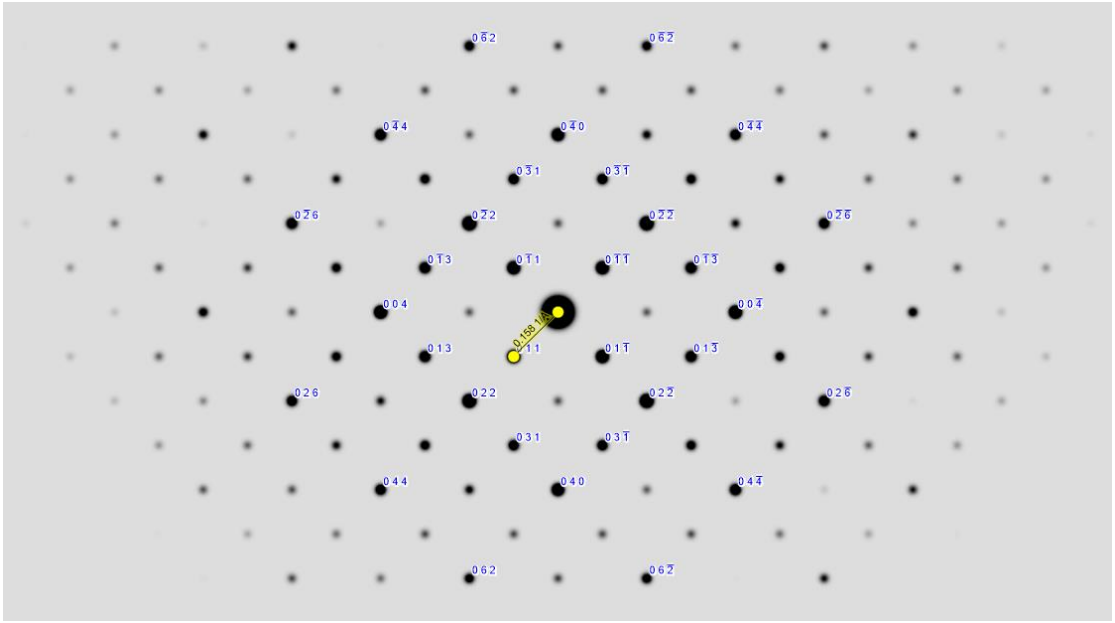

Figure S4 Simulated transmission electron diffraction of $FASnI_3$ single crystal from view direction of indices [100]. The simulation was performed by CrystalMaker and SingleCrystal Softwares according to the CIF of $FASnI_3$ (orthorhombic Amm2 space group).

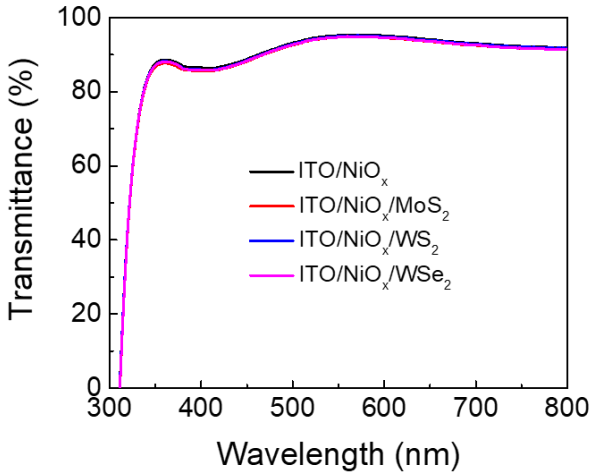

Figure S5 Transmittance spectra of NiO_x/ITO/glass substrates processed w/o and with MX_2 interlayer.

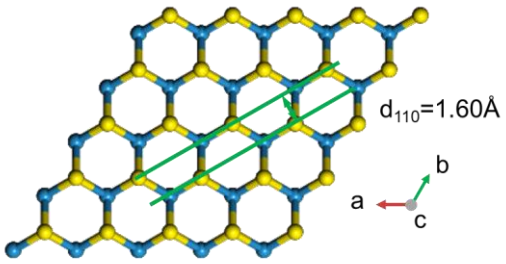

Figure S6 Atomic crystal structure of the (110) plane of WSe_2 from the top view.

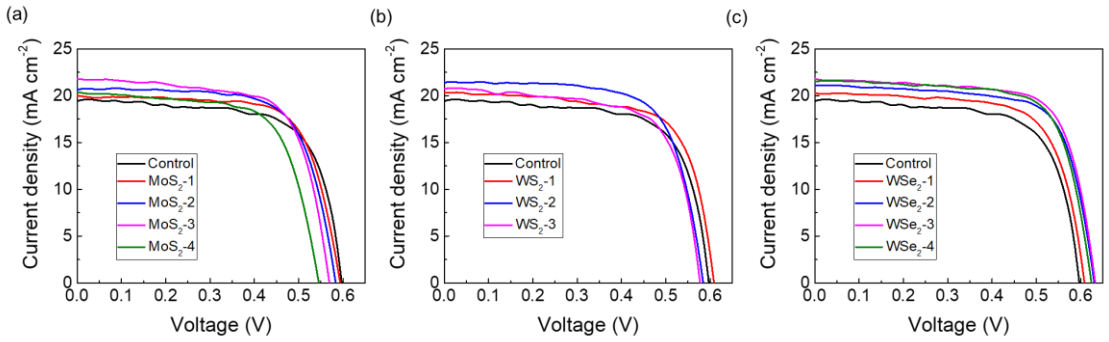

Figure S7 Typical J-V curves of the control PSC and MX_2- incorporated PSCs prepared by spin-coating MX_2 dispersions for different times.

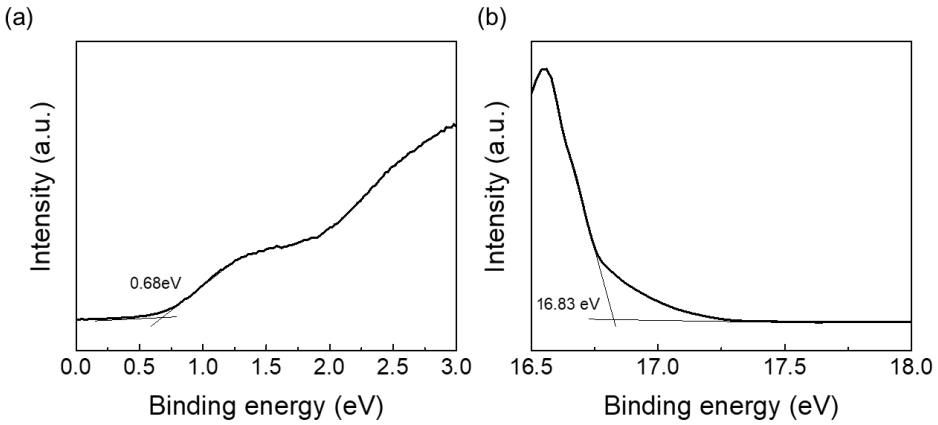

Figure S8 a) Near E_f region and b) the secondary electron cutoff region of UPS spectra of the $FASnI_3$ perovskite film.

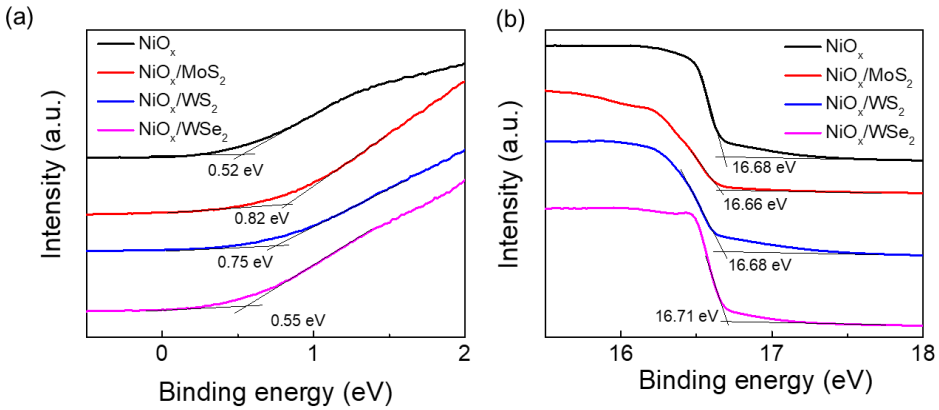

Figure S9 a) Near E_f region and b) the secondary electron cutoff region of UPS spectra of the NiO_x layer and MX_2 modified NiO_x layers.

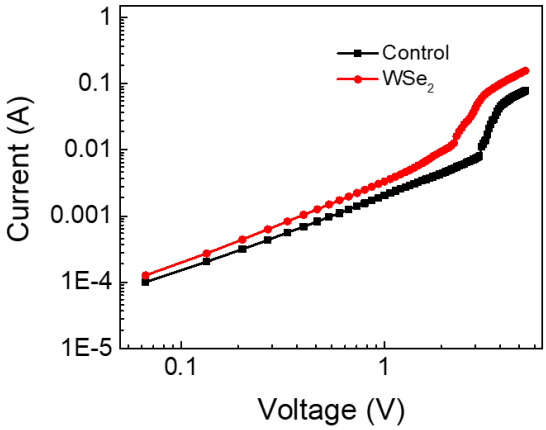

Figure S10 Dark I-V curves of the hole-only devices w/o and with WSe_2 incorporation.

Note: Hole-only devices with a structure of ITO/NiO_x/w/o or with WSe_2/$FASnI_3$/P3HT/Au were used to measure the dark I-V curves. The trap density (N_t) in the perovskite film can be calculated by the equation:

$$N_t = \frac{2\varepsilon\varepsilon_0 V_{TFL}}{qL^2}$$

where V_{TFL} is the trap filled limit voltage, ε is the relative dielectric constant of $FASnI_3$ (~5.7), ε_0 is the vacuum permittivity, q is the electron charge, and L is the perovskite film thickness.

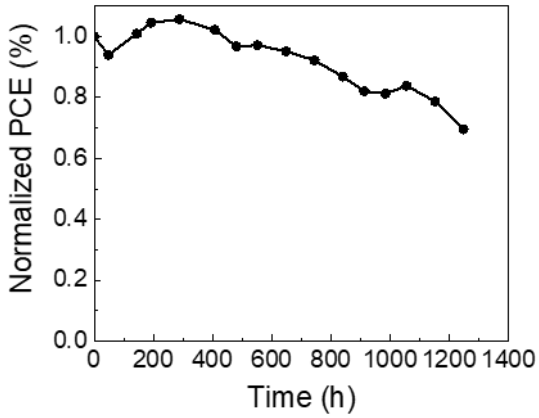

Figure S11 Long-term stability of the WSe_2 incorporated PSC (unencapsulated) when stored in the air with a relative humidity (RH) about 20% and measured in the ambient condition (RH≈45%).

Tables:

Table S1 Photovoltaic parameters of MX_2 processed PSCs (MX_2 dispersions were spin-coated for different times) extracted from Figure S7.

	V_{OC} (v)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
Control	0.59	19.44	70.0	8.03
MoS_2-1	0.59	20.02	71.0	8.39
MoS_2-2	0.58	20.67	70.4	8.44
MoS_2-3	0.57	21.75	68.7	8.52
MoS_2-4	0.54	20.40	67.8	7.47
WS_2-1	0.61	20.32	69.8	8.65
WS_2-2	0.58	21.44	70.4	8.75
WS_2-3	0.58	20.71	69.9	8.39
WSe_2-1	0.61	20.27	70.0	8.66
WSe_2-2	0.63	21.14	71.9	9.56
WSe_2-3	0.63	21.82	72.0	9.90
WSe_2-4	0.62	21.54	72.4	9.67

Table S2 Efficiencies of $FASnI_3$ –based PSCs in literature.

Composition	**Device structure**	**PCE (%)**	**Ref**
$FASnI_3$ (SnF_2)	FTO/c-TiO_2/mp-TiO_2/perovskite/spiro-OMeTAD/Au	2.1	[1]
$FASnI_3$ (SnF_2, N_2H_5Cl)	ITO/PEDOT:PSS/perovskite/PCBM/BCP/Ag	5.4	[2]
$FASnI_3$ (SnF_2)	ITO/PEDOT:PSS/perovskite/C_{60}/BCP/Ag	6.22	[3]
$FASnI_3$ (SnF_2, TMA)	ITO/PEDOT:PSS/perovskite/C_{60}/ bis-C_{60}/Ag	7.09	[4]
$FASnI_3$ (SnF_2, pyrazine)	FTO/c-TiO_2/mp-TiO_2/perovskite/spiro-OMeTAD/Au	4.8	[5]
$FASnI_3$ (SnF_2, $EDAI_2$)	ITO/PEDOT:PSS/ perovskite/C_{60}/BCP/Ag	8.9	[6]
$FASnI_3$ (SnF_2, PTN-Br)	ITO/PEDOT:PSS/perovskite/C_{60}/BCP/Ag	7.94	[7]
$FASnI_3$ (SnF_2,FOEI)	ITO/PEDOT:PSS/ perovskite/C_{60}/BCP/Ag	10.16	[8]
$FASnI_3$ (SnF_2, $EDAP_2$)	ITO/PEDOT:PSS/ perovskite/C_{60}/BCP/Ag	6.8	[9]
$FASnI_3$ (SnF_2, PHCl)	ITO/PEDOT:PSS/ perovskite/C_{60}/BCP/Ag	11.4	[10]
$FASnI_3$ (SnF_2, PAI)	ITO/PEDOT:PSS/ perovskite/C_{60}/BCP/Ag	11.22	[11]
$FASnI_3$ ($SnCl_2$,GA)	ITO/NiO_x/WSe_2/perovskite/PCBM/BCP/Ag	10.47	This work

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