

Supplementary Information

S1 Fabrication of photodetectors

A. Silicon wafer surface cleaning

We use decontamination powder to scrub the silicon wafer, and then use decon90 and isopropanol to ultrasonically clean it for 10 minutes in sequence. Finally, we rinse the silicon wafer with deionized water and dry it with nitrogen.

B. Patterned electrode preparation

First, the photoresist was spin-coated on the surface of the substrate. The spin coating procedure was divided into two steps: the first step took 8 seconds and the rotation speed was 1000 rpm; the second step took 52 seconds and the rotation speed was 3000 rpm. The spin-coated sample was placed on a hot plate for pre-baking at a temperature of 120° C for 60 seconds. Then routine photolithography was performed, and the exposed sample was developed after being heated at 120°C for 90 seconds. After the development is completed, the silicon wafer is rinsed and dried, and then heated on a hot plate for 120 seconds. Then the electrodes were prepared, the evaporation rate was 3 Å/s, and the total thickness of the evaporation was 100 nm.

Finally, soak the silicon wafer with electrodes steamed in degumming solution, place it in a water bath at 60°C and heat it for 15 minutes, rinse it in deionized water after degumming, and then carefully rinse with deionized water and blow dry. The dried electrodes were heated and dried on a hot plate.

C. Preparation of Zn²⁺ doped CsPbCl₃ perovskite film

The perovskite thin film was fabricated using a three-source vacuum evaporation method. After setting the material evaporation temperatures, the heating current was gradually increased to stabilize the evaporation rates of cesium chloride (CsCl) and lead chloride (PbCl₂) at 1.5 Å/s. For zinc chloride (ZnCl₂) with different doping ratios (e.g. 11% doping required precise rate control at 0.37 Å/s), the corresponding evaporation rates were carefully regulated. The evaporation process was terminated once the desired thickness was achieved. Following evaporation, the perovskite film underwent vacuum annealing at 75°C for 10 minutes.

Finally, a C₆₀ layer was thermally deposited at a rate of 1 Å/s to complete the device structure.

S2 Calculation of composition of ZnCl₂ doped CsPbCl₃ based on EDS spectra

Sample	Atomic Fraction (%)			
	Cs	Pb	Zn	Cl
CsPbCl ₃	22.89	20.33	0	56.78
CsPbCl ₃ :4%Zn	19.12	17.26	1.51	62.11
CsPbCl ₃ :7.6%Zn	18.68	14.10	2.72	64.50
CsPbCl ₃ :11%Zn	17.19	13.53	3.81	65.47
CsPbCl ₃ :14.5%Zn	17.33	13.45	5.21	64.01

Taking 11% Zn²⁺ doping as an example, the EDS ratios of different elements are as follows.

Cs=17.19 Pb=13.53 Zn=3.81 Cl=65.47

So the doping ratio of Zn^{2+} is: $\frac{3.81}{17.19 + 13.53 + 3.81} \approx 0.11 = 11\%$

S3 Derivation of the PL decay life time by the exponential fitting of TRPL decaying curves

The calculation formula for the average fluorescence lifetime of the three index fitting fluorescence lifetime is based on the weighted average of the amplitude (A_i) and lifetime (T_i) of each index component, where the weight is the integral contribution of each component. The specific formula is as follows:

$$T_{average} = \frac{(A_1 T_1^2 + A_2 T_2^2 + A_3 T_3^2)}{(A_1 T_1 + A_2 T_2 + A_3 T_3)}$$

Taking 11% Zn^{2+} doping sample as an example:

$$T_{average} = \frac{(0.35 \times 0.59^2 + 0.06 \times 2.27^2 + 0.47 \times 13.69^2)}{(0.35 \times 0.59 + 0.06 \times 2.27 + 0.47 \times 13.69)} = 1.839(ns)$$

S4 Calculation of the noise equivalent power (NEP) and specific detectivity (D^*)

The parameter of detectivity (D) is the signal detecting sensitivity of a PD from noise environment, which is defined as the inverse of the noise equivalent power (NEP). When the NEP is normalized by the square root of the product of the detector area (S), bandwidth (Δf), and the dark current (I_d), its reciprocal is defined as the D^* . The unit of the D^* is in Jones ($cm \text{ Hz}^{1/2} W^{-1}$). In the circumstances where the shot noise is the dominant contributor, the D^* can be calculated as follows:

$$D^*(Jones) = \frac{(S \Delta f)^{1/2}}{NEP} = \frac{R_\lambda}{(2eI_d / S)^{1/2}}$$

$$NEP = \frac{(S \Delta f)^{1/2}}{D^*(Jones)}$$

Taking the pristine $CsPbCl_3$ sample at the λ of 500 nm as an example:

$$D^* = \frac{2060 A/W}{(2 \times 1.6 \times 10^{-19} C \times 2.1 \times 10^{-6} A / (18 \times 10^{-4} cm \times 100 \times 10^{-4} cm))^{1/2}} = 1.07 \times 10^{13} cmHz^{1/2} W^{-1}$$

$$NEP = \frac{(18 \times 10^{-4} cm \times 100 \times 10^{-4} cm \times 5.04 \times 10^{14} Hz)^{1/2}}{1.07 \times 10^{13} cmHz^{1/2} W^{-1}} = 8.16 \times 10^{-9} W$$

S5 Calculation of defects density and carrier mobility based on SCLC mode

The trap density of the $CsPbCl_3$ is derived by the capacitance (C)–voltage(V) measurement of the perovskite device based on equation 1, with the device structure of ITO/ MnO_3 (2nm)/ $CsPbCl_3$ /Ag (80nm):

$$N_t = \frac{2\epsilon\epsilon_0 V_{TFL}}{eL^2} \quad (1)$$

Taking the calculation of defect density in the 11% Zn^{2+} doped sample as an example:

$$N_t = \frac{2 \times 1.13 \times 8.85 \times 10^{-12} Fm^{-1} \times 0.53V}{1.61 \times 10^{-19} C \times 1.09 \times 10^{-7} m \times 1.09 \times 10^{-7} m} = 0.55 \times 10^{16} cm^{-3}$$

in which the V_{TFL} stands for the onset voltage of the trap fill limit regime while ϵ , ϵ_0 , e and L represent the relative dielectric constant of the films, the vacuum permittivity, elementary charge, and perovskite thickness, respectively. The obtained capacitance(C)–voltage(V) curves and V_{TFL}

are shown in Fig.S4 in the support information and the calculated trap density of pristine and Zn with C₆₀ capped CsPbCl₃ is listed in Table 3.

To study the carrier mobility after passivation of CsPbCl₃ with Zn and C₆₀, based on the space-charge-limited current (SCLC) model, namely, the Mott-Gurney law, the mobility can be derived by equation 2:

$$J = \frac{9\epsilon\epsilon_0\mu V^2}{8L^3} \quad (2)$$

Taking the carrier mobility in the 11% Zn²⁺ doped sample as an example:

$$\mu = \frac{8 \times 692.5 A / m^2 V^2 \times (1.09 \times 10^{-7})^3 m^3}{9 \times 1.13 \times 8.85 \times 10^{-12} F m^{-1}} = 7.971 \times 10^{-4} cm^2 / Vs$$

The dependence of the carrier mobility on the applied voltage can be derived accordingly. The perovskite film has bipolar nature and nearly the same electron and hole mobility, so we can obtain electron mobility by fitting I-V curve according to the equation 2 where μ , V, L stand for the carrier mobility, applied voltage and thickness of the perovskite, respectively, and the results are shown in Table 4

S6 Calculation of the electronic structure (E_c, E_v and E_f)

UPS is used to determine the Fermi level (E_F) and the valence band maximum (E_V) with respect to vacuum level (E_{VAC}) of the CsPbCl₃ thin films.

For a photoelectron to escape the sample surface and to be collected, it has to have sufficient energy to overcome the sum of the binding energy (with respect to E_F) of its initial level and the work function (Φ), where $\Phi = E_{VAC} - E_F$. Therefore, for a fixed incident photon energy of 21.2eV, the secondary electron cut-off (high binding energy edge) represents photoelectrons with zero kinetic energy (E_k) when they escape the sample surface. The work function Φ is determined by the difference between the incident photon energy (21.2eV) and the binding energy of the secondary electron cut-off. In the example spectrum, the cut-off binding energy is 16.9eV as determined by the intersection of the linear portion of the spectrum and the baseline. The work function of this sample is thus $\Phi = 21.2 - 16.9 = 4.32$ eV; that is, E_F is -4.32eV with respect to E_{VAC}. The example spectrum has a E_F-E_V=1.47eV. Therefore, its valence band maximum E_V is -5.8eV with respect to E_{VAC}.

Table S1 Surface roughness and thickness

Sample	RMS roughness(nm)	Mean roughness(nm)	Thickness(nm)
CsPbCl ₃	17.09	14.26	99.3
CsPbCl ₃ :4.0%ZnCl ₂	8.48	5.17	102.5
CsPbCl ₃ :7.6%ZnCl ₂	3.92	2.46	104.7
CsPbCl ₃ :11.0%ZnCl ₂	2.17	1.74	106.2
CsPbCl ₃ :14.5%ZnCl ₂	7.82	6.39	111.2
CsPbCl ₃ :11.0%ZnCl ₂ +5nmC ₆₀	5.78	4.41	109.7
CsPbCl ₃ :11.0%ZnCl ₂ +10nmC ₆₀	2.45	1.96	113.6
0			
CsPbCl ₃ :11.0%ZnCl ₂ +15nmC ₆₀	2.50	2.16	117.8
0			

Table S2 Grain size calculation based on Scherrer equation

	Peak position (2 Theta)	FWHM (2 Theta)	Crystal size D (nm)	D average (nm)
CsPbCl ₃	15.741	0.194	40.9	32.9
	22.413	0.208	38.5	
	31.821	0.297	27.5	
	35.250	0.246	33.5	
	50.655	0.361	24.1	
CsPbCl ₃ :4.0%ZnCl ₂	15.729	0.218	36.4	32.6
	22.453	0.242	33.1	
	31.961	0.273	29.9	
	35.235	0.224	36.8	
	50.674	0.327	26.6	
CsPbCl ₃ :7.6%ZnCl ₂	15.808	0.247	32.1	30.9
	22.492	0.234	34.2	
	31.998	0.307	26.6	
	35.271	0.230	36.0	
	50.675	0.339	25.6	
CsPbCl ₃ :11.0%ZnCl ₂	15.846	0.280	28.3	30.0
	22.473	0.221	36.2	
	31.999	0.273	29.9	
	35.233	0.264	31.2	
	50.635	0.357	24.3	
CsPbCl ₃ :14.6%ZnCl ₂	15.828	0.273	29.1	24.3
	22.473	0.287	27.9	
	32.019	0.428	19.1	
	39.378	0.343	24.3	

	51.501	0.411	21.2	
	15.863	0.226	35.1	
	22.473	0.196	40.9	
CsPbCl ₃ :11.0%ZnCl ₂ +5nmC ₆₀	31.975	0.231	35.4	34.2
	35.274	0.233	35.4	
	50.673	0.361	24.2	
	15.887	0.217	36.6	
	22.513	0.220	36.4	
CsPbCl ₃ :11.0%ZnCl ₂ +10nmC ₆₀	32.065	0.289	28.3	32.2
	35.274	0.233	35.4	
	50.694	0.355	24.5	
	15.786	0.250	31.7	
	22.475	0.210	38.1	
CsPbCl ₃ :11.0%ZnCl ₂ +15nmC ₆₀	31.921	0.204	40.1	33.3
	35.310	0.248	33.2	
	50.676	0.374	23.2	

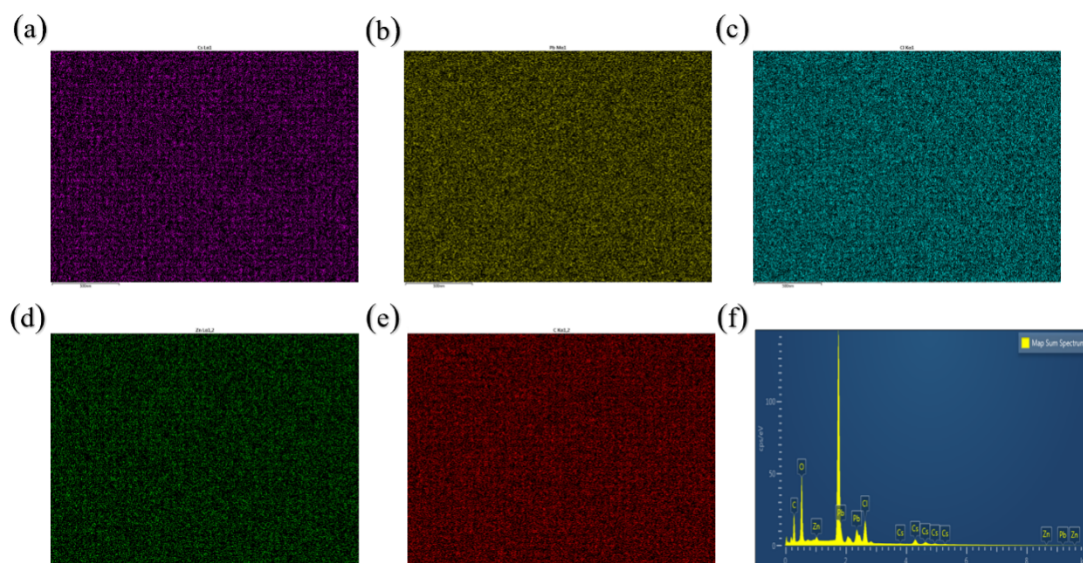


Fig.S1 EDS mapping analysis of sample 10C-11Z.

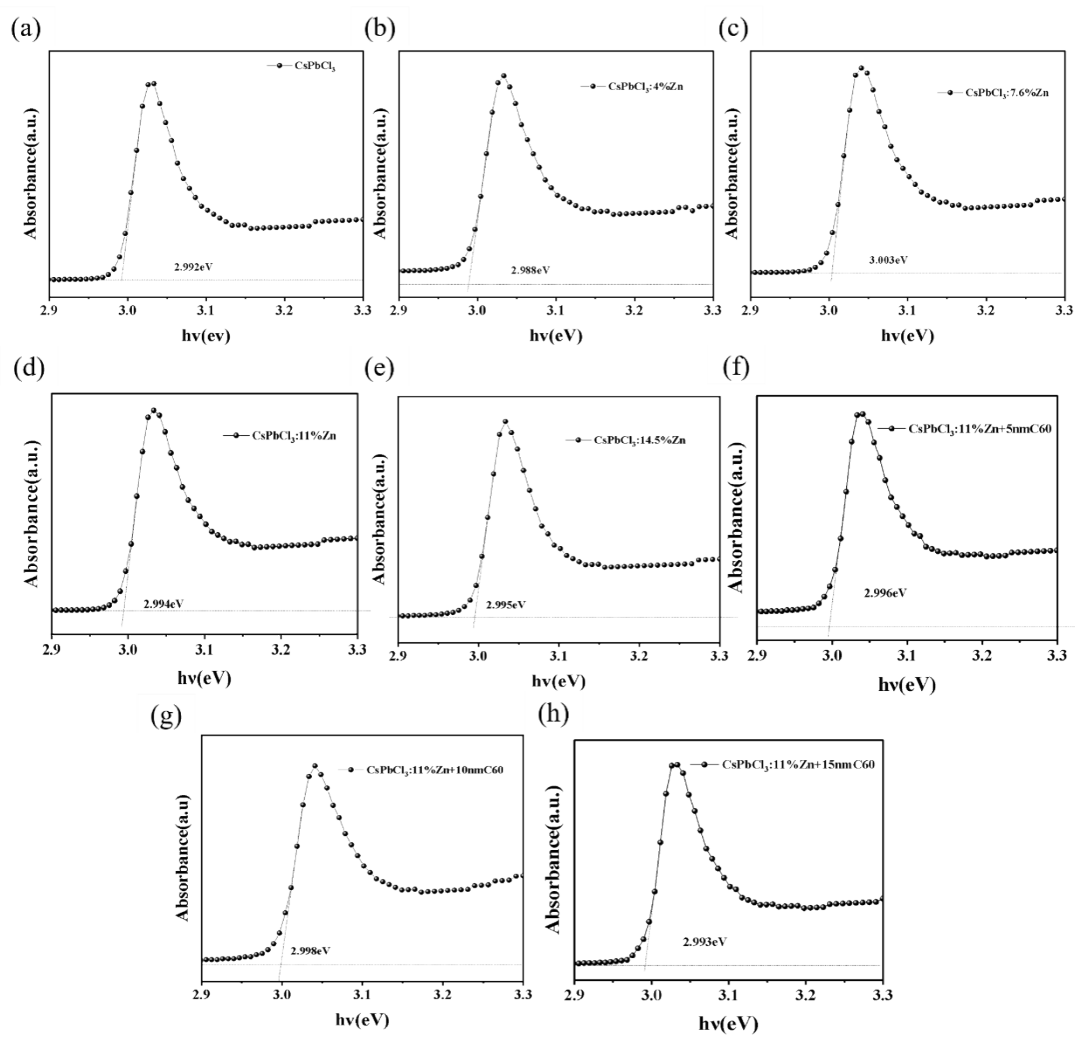
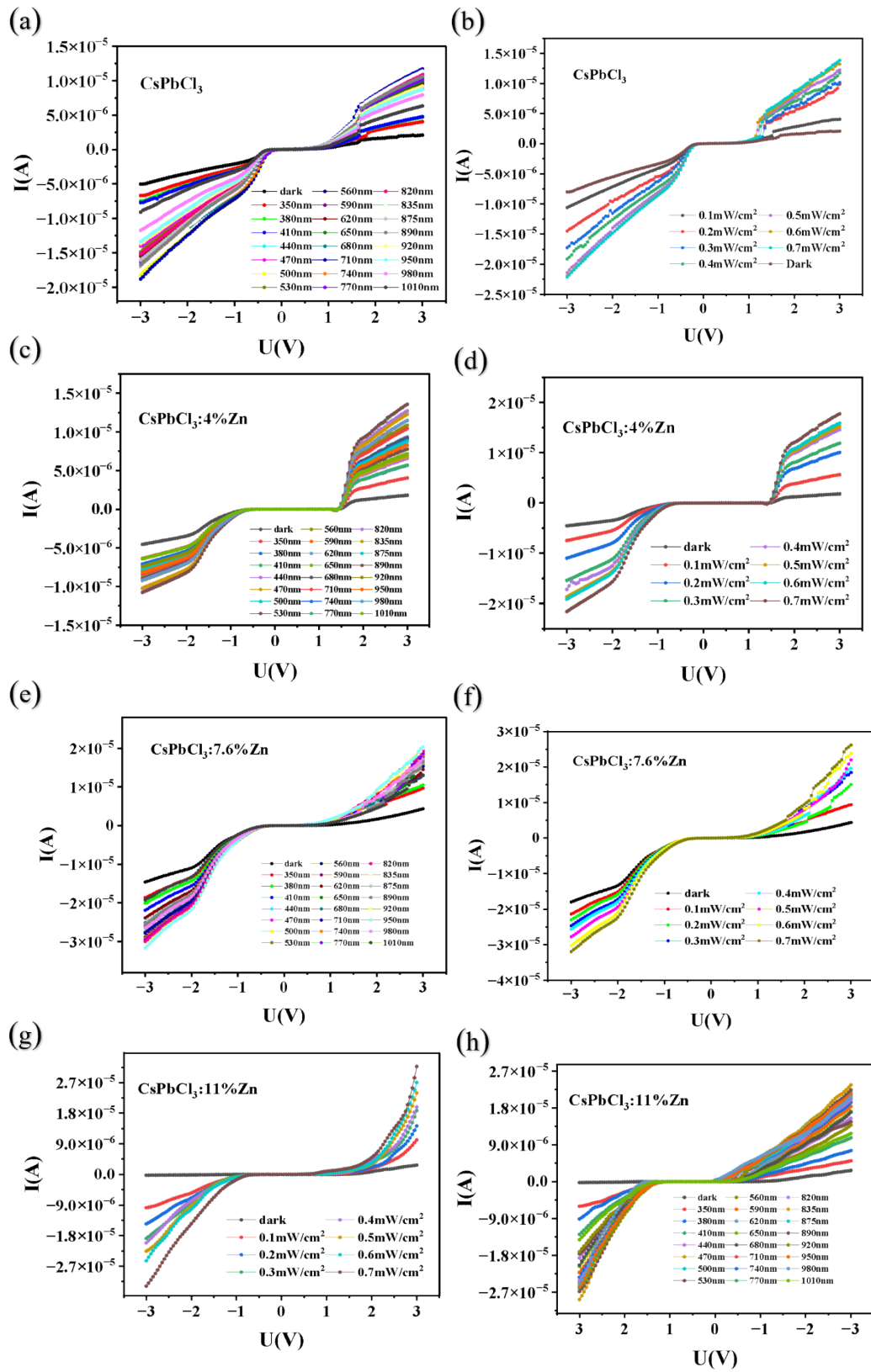


Fig.S2 Taucs plot for all samples



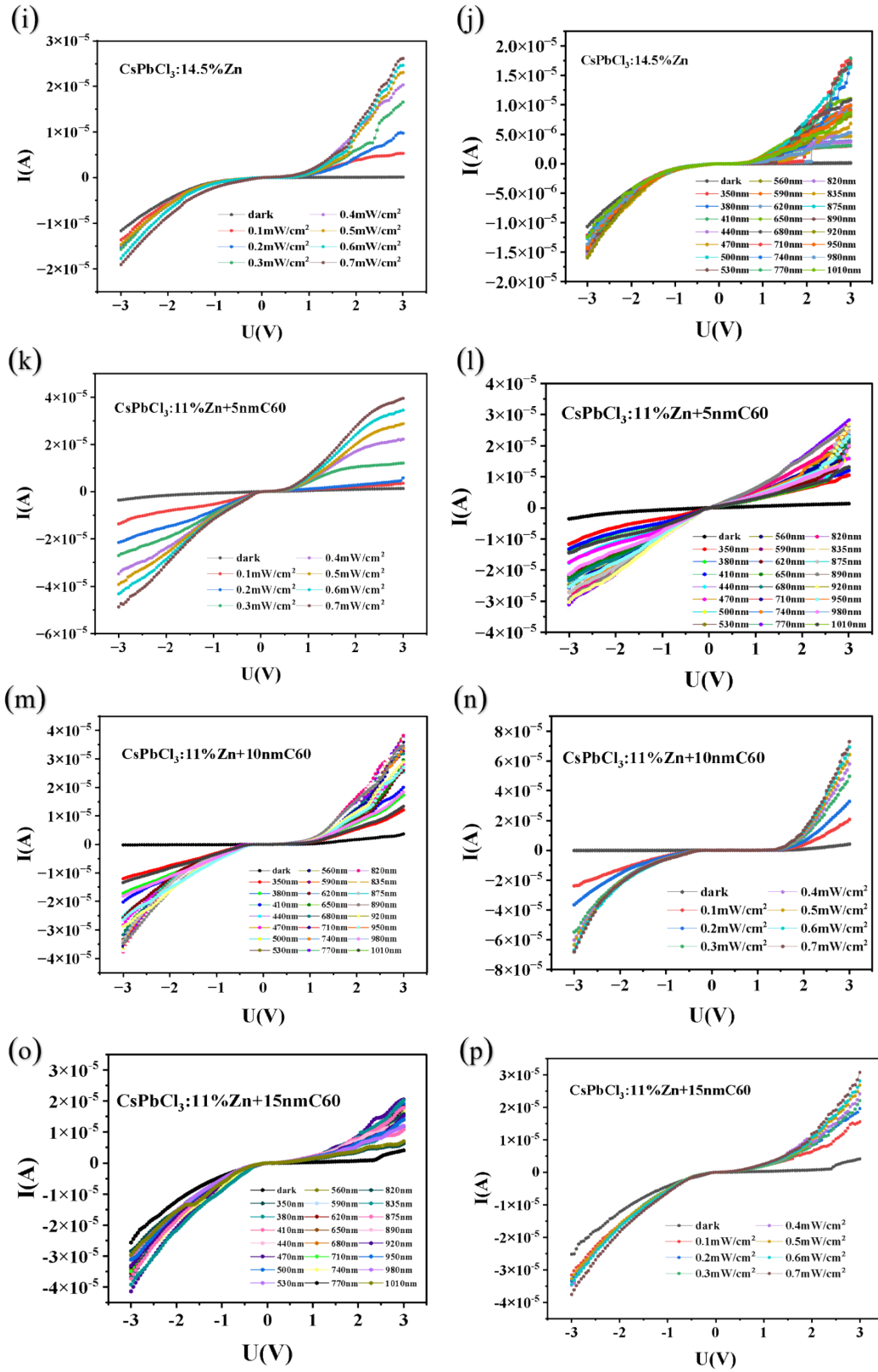


Fig.S3 Photocurrent (I)-applied voltage (U) curves at fixed incident $\lambda = 500 \text{ nm}$, $U = 3 \text{ V}$ with various light power and at fixed incident light power of 0.2 mW/cm^2 at various λ for sample P (a,b), 4Z (c,d), 7.6Z (e,f), 11Z (g,h), 14.5Z (i,j), 5C-11Z (k,l), 10C-11Z (m,n), 15C-11Z (o,p)

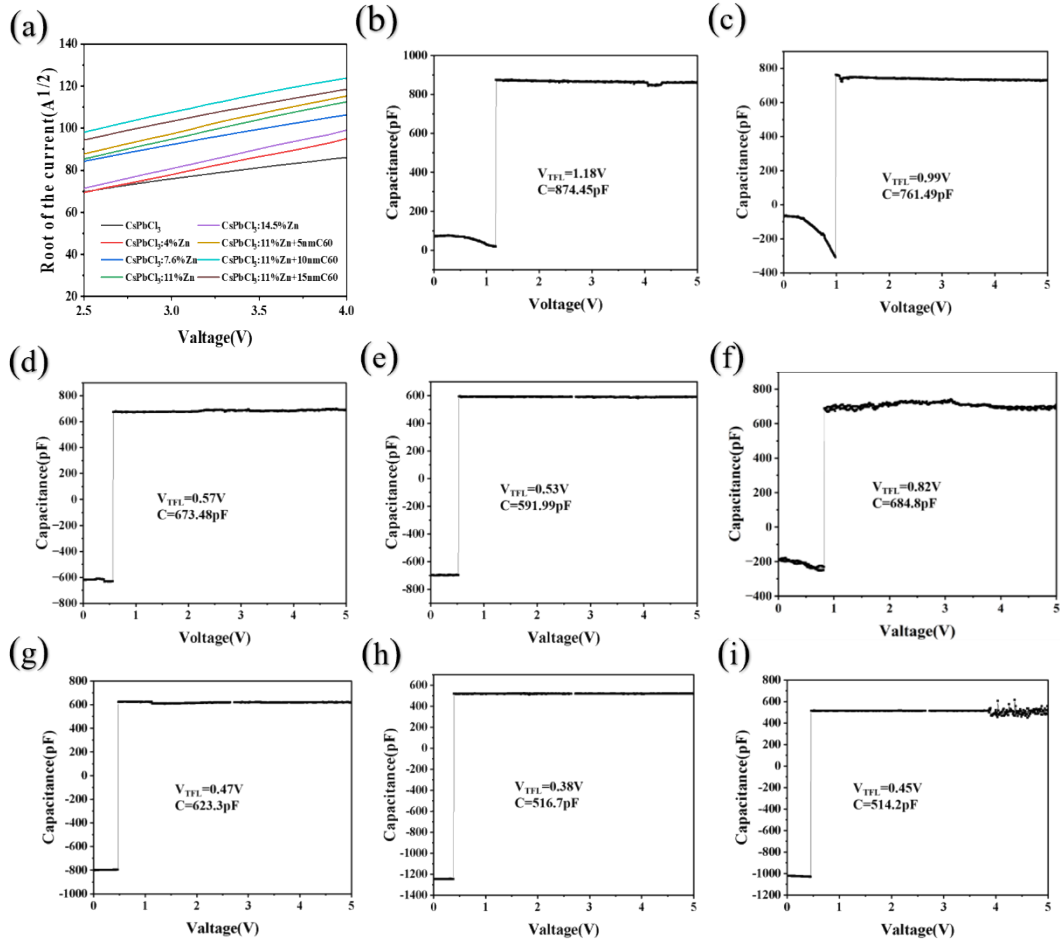


Fig.S4 I-V(a) and C-V(b-i) curves based on SCLC mode of all samples