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Polymer-Assisted Crystal Growth Regulation and Defect Passivation for High-Performance Flexible Solar-blind Photodetector based on Copper-Based Halides

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Experiment Section

Material:

CsI (≥99.9%), CuI (≥99.999%), poly(vinylpyrrolidone) (PVP) (M_W ~ 40000), poly(4-vinylphenol), polyethylene oxide (PEO) and polystyrene (PS) were purchased from Sigma-Aldrich. Dimethyl sulfoxide (≥99.7%), N, N-Dimethylformamide (≥99.8%) and toluene (Analytical Reagent) were purchased from Meryer. All the chemicals were used without further purification.

Preparation of Polymer-$Cs_3Cu_2I_5$ Precursor Solution:

0.75 M $Cs_3Cu_2I_5$ precursor was prepared by dissolving CsI and CuI into a mixture of DMF and DMSO (1:1 v/v). The mixture was stirred at 70 ℃ for 12 h until completely dissolved. Thereafter, a colorless precursor solution was obtained by filtering the crude solution through a PTFE filter (0.45 μm). Different amount of polymers was added into the precursor solution to prepare PVP- $Cs_3Cu_2I_5$ precursor solution.

Preparation of Polymer-$Cs_3Cu_2I_5$ Device:

Firstly, the 100 mL as-prepared precursor solution was spin-coated on the quartz substrates in the argon-filled glovebox, involving a low-speed (500 rpm, 5 s) and high-speed (3000 rpm, 60 s) spinning process. A wet layer of Polymer-$Cs_3Cu_2I_5$ film was formed in the first 45 seconds. Then, 100 μL of toluene as an anti-solvent was dropped onto the top of the spinning layer to promote a fast crystallization for film formation. Finally, the sample was annealed on a hotplate at 100°C for 5 min to evaporate the solvent and form the compact Polymer-$Cs_3Cu_2I_5$ films.

Characterizations:

PL, PL excitation (PLE), PLQY, and time-resolved PL (TRPL) spectra were measured via an Edinburgh instruments FS5 spectrometer equipped with an integrating sphere. The chemical compositions and film morphology were analyzed with an energy-dispersive spectrometer (EDS)-equipped field-emission scanning electron microscope (SEM) JEOL-7800F. X-ray diffraction (XRD) measurement was carried out using a Bruker D8 Advance. Responsivity and I−V curve was measured by a Keithley 2400 semiconductor analyzer, and the light source with continuously changing wavelength is provided by a Zolix tunable 500-W arc lamplight.

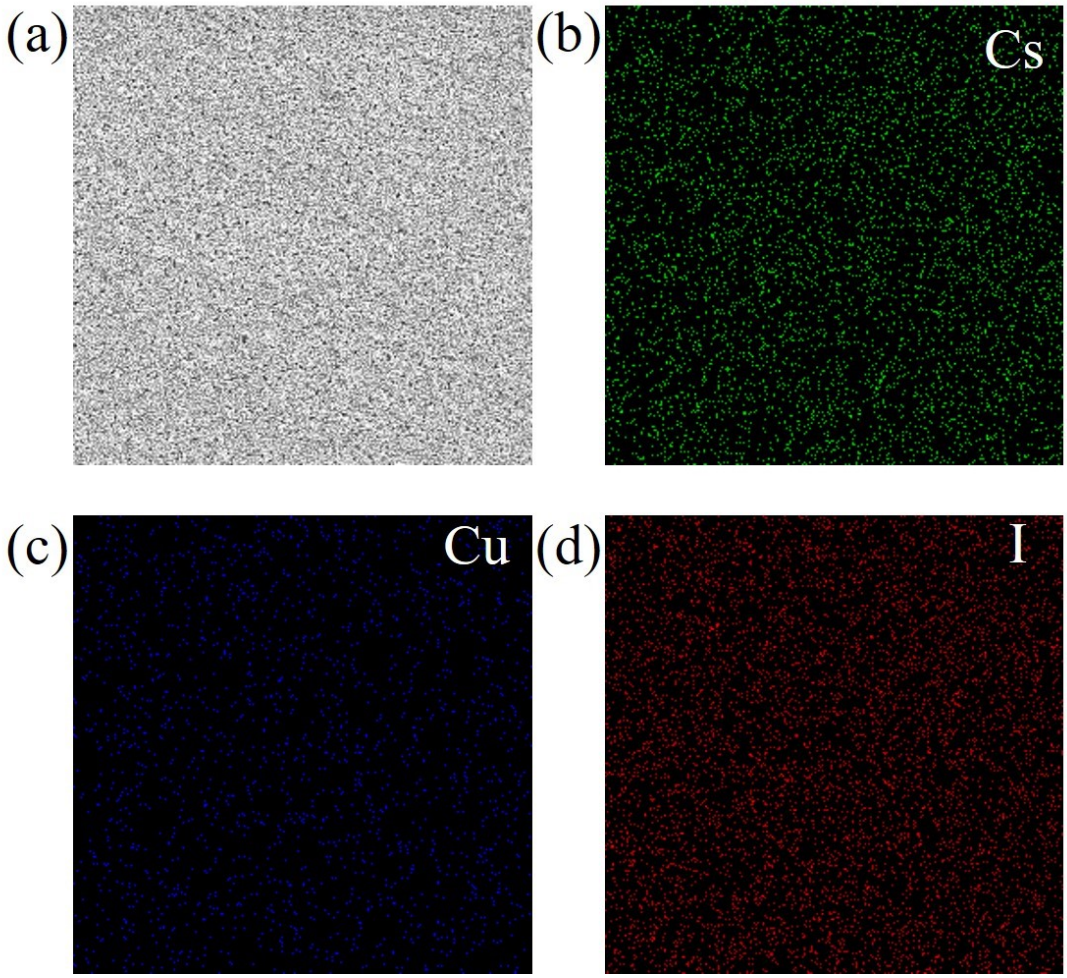

Figure S1. (a) SEM image of $Cs_3Cu_2I_5$ thin films. (b-d) EDS results of $Cs_3Cu_2I_5$ film.

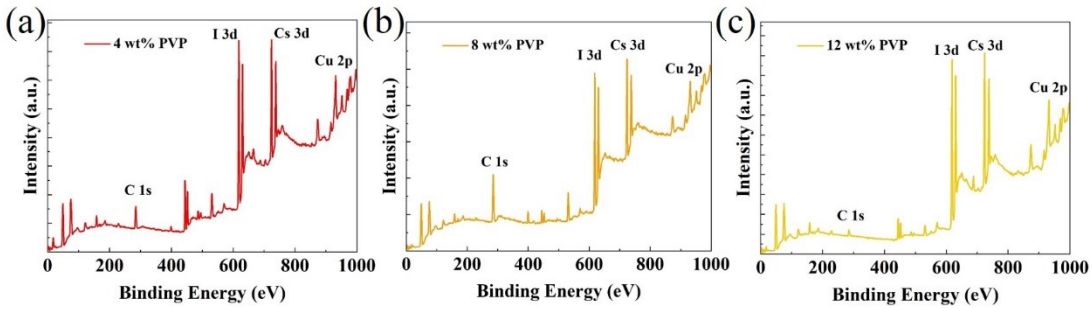

Figure S2. Total XPS spectrum of the prepared (a) 4, (b) 8 and (c) 12 wt% PVP-$Cs_3Cu_2I_5$ film.

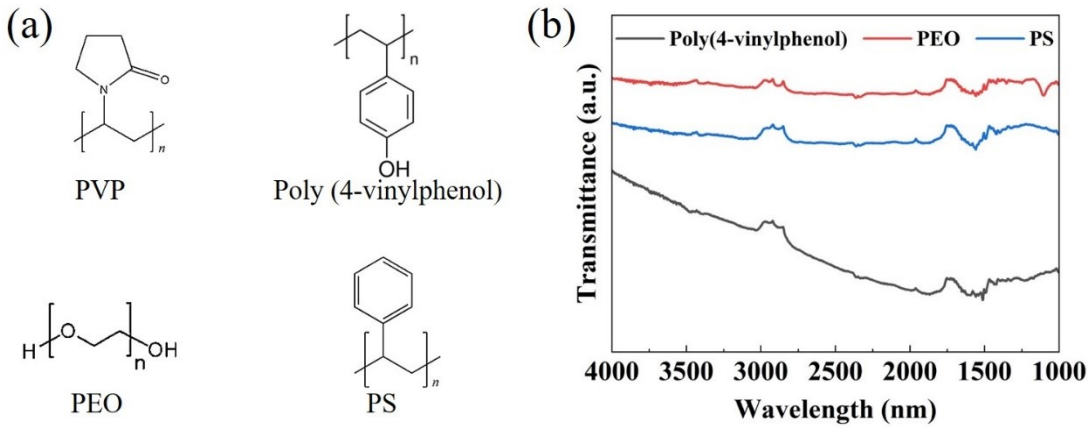

Figure S3. (a) Structure of different polymers. (b) FT-IR spectra of $Cs_3Cu_2I_5$ film with different polymers.

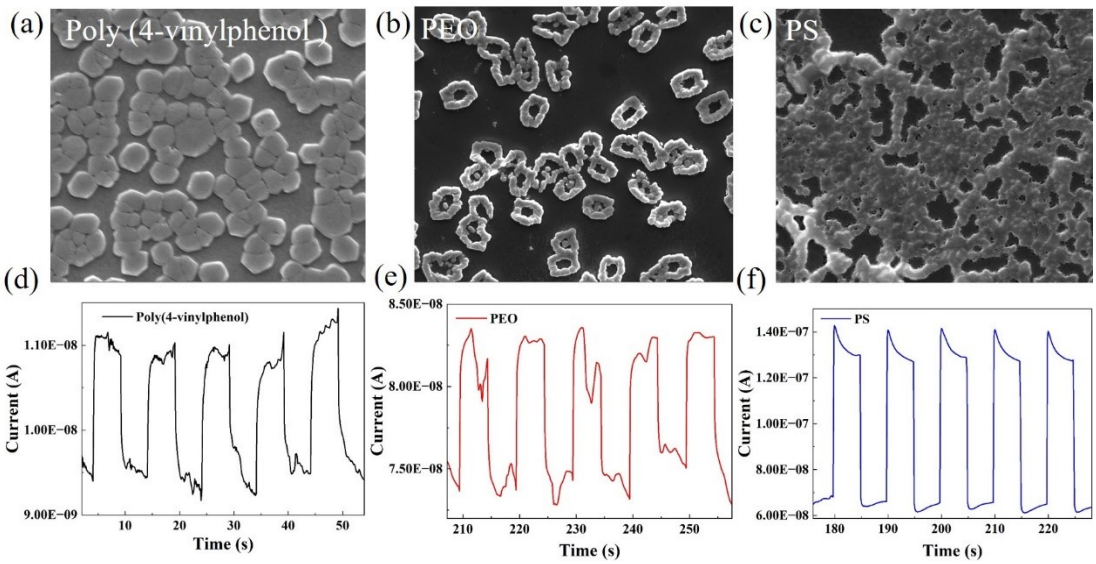

Figure S4. (a-c) SEM spectra of $Cs_3Cu_2I_5$ with different polymers film. (d-f) Response curve of $Cs_3Cu_2I_5$ with different polymers film.

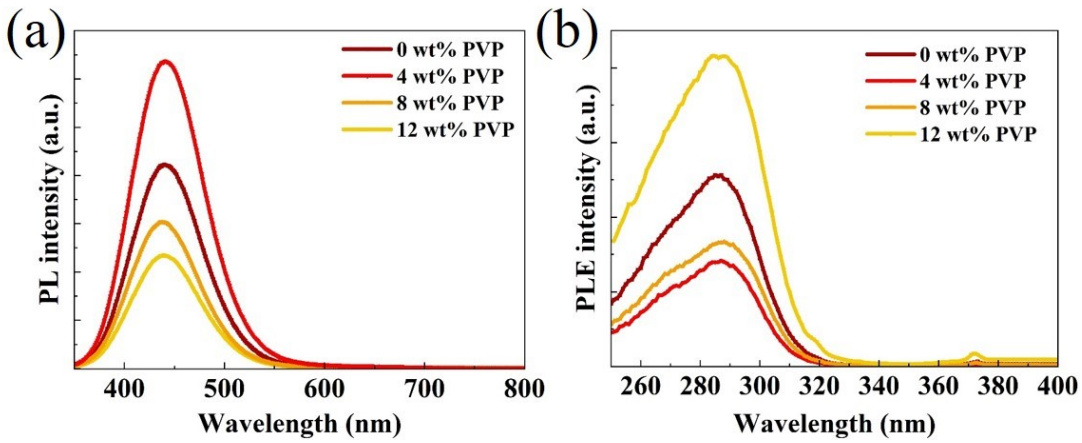

Figure S5. (a) PL spectrum, (b) PLE spectrum of 0, 4, 8 and 12 wt% PVP-$Cs_3Cu_2I_5$ films.

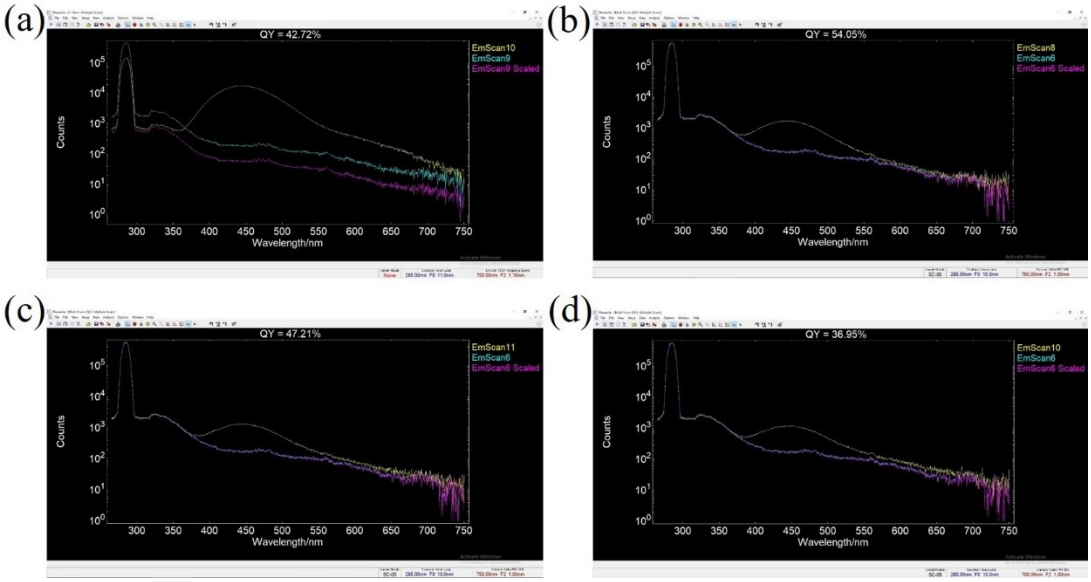

Figure S6. PLQY of the prepared (a) 0, (b) 4, (c) 8 and (d) 12 wt% PVP-$Cs_3Cu_2I_5$ film.

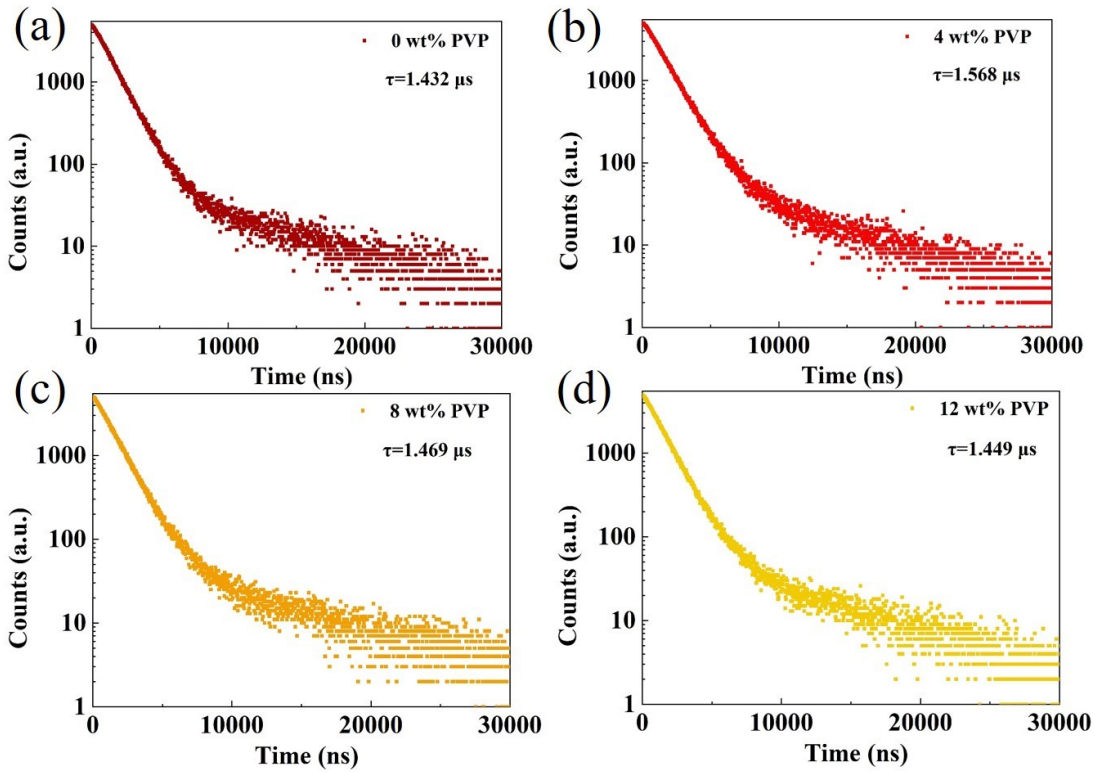

Figure S7. TRPL of the prepared (a) 0, (b) 4, (c) 8 and (d) 12 wt% PVP-$Cs_3Cu_2I_5$ film.

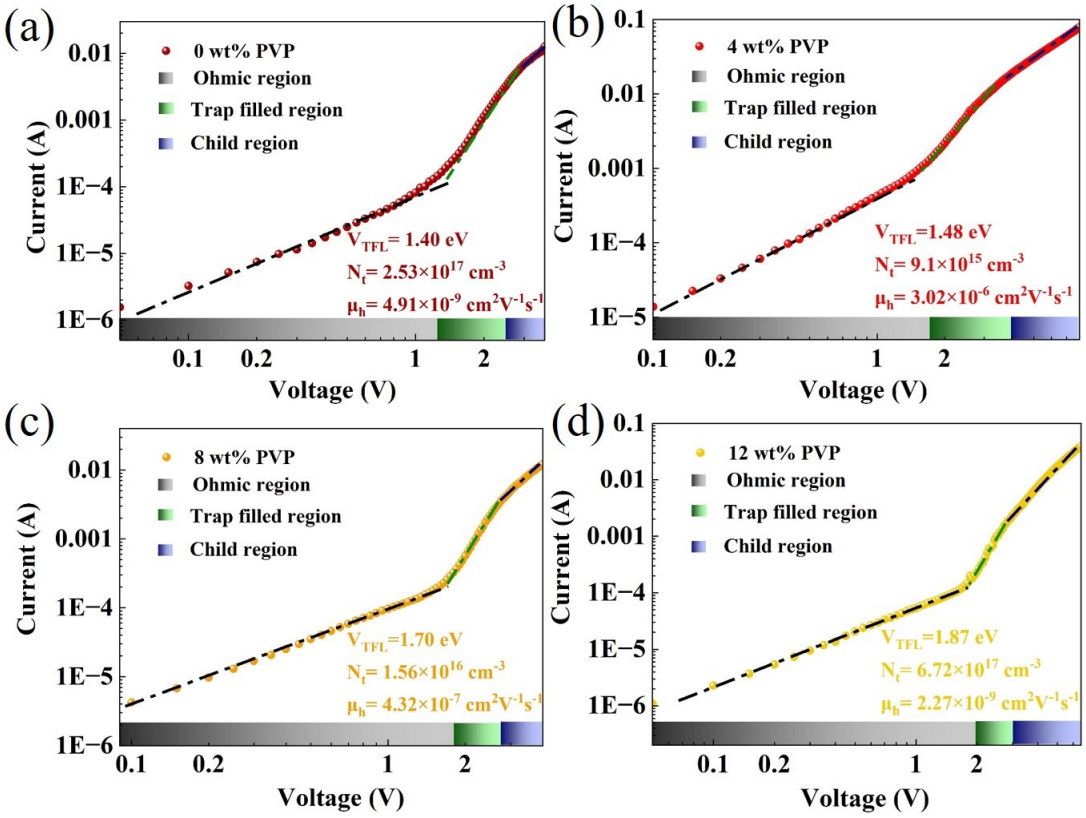

Figure S8. Double logarithmic current density–voltage curves of the hole-only devices based on (a) 0, (b) 4, (c) 8 and (d) 12 wt% PVP-$Cs_3Cu_2I_5$ films.

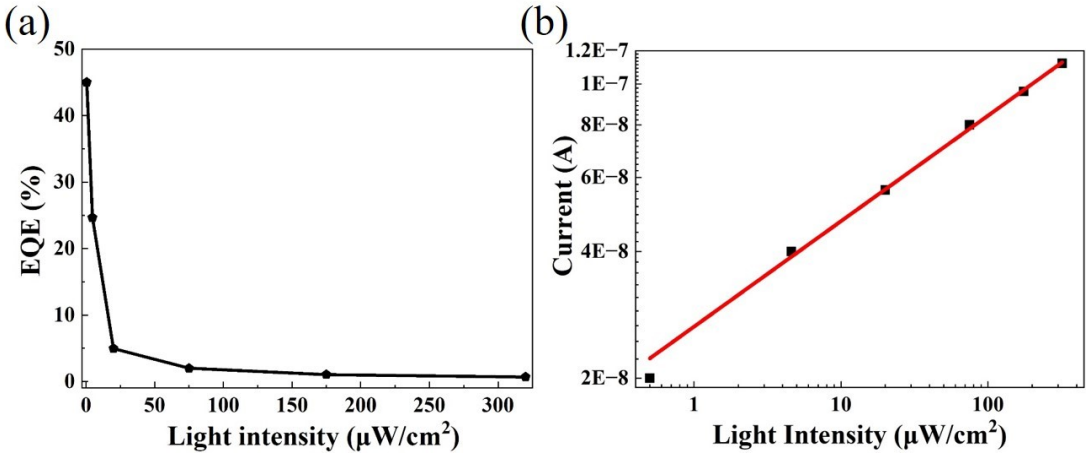

Figure S9. (a) Dependence of EQE upon intensity (bias voltage=1 V). (b) Logarithmic plot of the photocurrent versus light intensity.

Table S1. Summary on copper-based halides photodetector.

Materials	Range of response	R	D*	Reference
$Cs_3Cu_2I_5$@PVP film	**200-263 nm**	**0.5 A/W**	**1.85×10^{12} Jones**	**This work**
$Cs_3Cu_2I_5$ film	200-300 nm	17.8 A/W	1.12×10^{12} Jones	[1]
$Cs_3Cu_2I_5$ crystalline film	200-380 nm	0.0649 A/W	6.9×10^{11} Jones	[2]
$Cs_3Cu_2I_5$/β-Ga_2O_3	200-280 nm	0.0023 mA/W	10^8 Jones	[3]
$Cs_3Cu_2I_5$/GaN	300-370 nm	0.28 A/W	1.4×10^{12} Jones	[4]
$Cs_3Cu_2Br_5$/GaN	200-275 and 325-375 nm	0.005 mA/W	5.29×10^{11} Jones	[5]
$CsCu_2I_3$	200-360 nm	0.047 mA/W	2.58×10^{12} Jones	[6]
$Cs_3Cu_2Br_5$/ZnO	200-380 nm	0.3 A/W	1.26×10^{11} Jones	[7]

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