

Supporting Information

Wavelength-Sensitive CMOS-like Optoelectronic Inverter Circuits Based on Solution-Processable Perovskite nanocrystals/Organic Semiconductor Blends

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Experimental Section & Analytical Procedures

Materials: The organic semiconductors were purchased from Derthon (P(NDI2OD-T2)) and 1-Material (PDVT-10).

Synthesis of CsPbX₃ (X = Br or I) NCs:

Preparation of Cs-oleate: Cs₂CO₃ (0.814g, Heowns, 99%) was taken into 100 ml 3-neck flask along with 1-Octadecene (40ml, ODE, Aladdin, 90%) and oleic acid (2.5 mL, OA, Heowns, 90%), dried for 1 hour at 120 °C under vacuum, then heated under Ar to 150°C until all Cs₂CO₃ reacted with oleic acid. Since Cs-oleate precipitates out of octadecene at room-temperature, it has to be preheated to ~100 °C before injection.

Synthesis of CsPbX₃ NCs: Octadecene (5 ml) and PbX₂ (0.188 mmol) such as PbI₂ (0.087g, Aladdin, 99.999%) or PbBr₂ (0.069g, Aladdin, 99.9%), were loaded into 50 mL 3-neck flask and dried for 1 hour at 120°C under vacuum. Dried oleylamine (0.5mL, OLAm, Heowns, 95%) and dried oleic acid (OA, 0.5 mL) were injected at 120 °C under Ar. Now temperature was raised to 140-200°C (for tuning the NC size) and Cs-oleate solution (0.4 mL, 0.125 M in ODE, prepared as described above) was quickly injected, and after 5s, the reaction mixture was cooled by the ice-water bath.

Isolation and purification of CsPbX₃ NCs. The combined NCs were separated by centrifuging after the crude solution was cooled using a water bath. For smaller NCs synthesized below 160 °C, centrifugation with the addition of dry methyl acetate (Damas-Beta, 99%) to the crude solution (ODE:Methyl acetate = 1:1 by volume) was shown to be beneficial for a complete precipitation. After centrifugation, the particles were re-dispersed in hexane or toluene to create long-term colloiddally stable solutions, and the supernatant was disposed of.

Device Fabrication: In this work, interdigitated bottom-contact bottom-gate configuration organic phototransistors were fabricated. Highly doped silicon wafers with 300 nm of thermally grown SiO₂ as the gate dielectric. Standard lithography procedures were used to pattern the wafers with Au electrodes of 50 nm using 10 nm of Ti as an adhesion layer. The channel length(L) and the width(W) of fabricated devices are 20 μm and 10 mm respectively. The

substrates were ultrasonically cleaned in acetone, isopropyl alcohol each for 30 min, and blown dry by nitrogen gas, and further treated by UV-ozone in the air for 5 min. The octadecyltrichlorosilane (OTS)-treated Si/SiO₂ substrates were used to suppress charge trapping effect between semiconductor/dielectric (see below). Samples with organic semiconductors in the channel were fabricated by spin-coating of the semiconductor or semiconductor /perovskite solution at a total concentration of 5 mg/mL, at 4000 rpm for 60s. All the solutions were prepared using chloroform as solvent. All solutions, samples and devices were prepared and measured in a nitrogen filled glovebox to avoid oxidative doping and degradation of the materials and ensure reproducibility of the experiments.

OTS treatment consisted of an overnight immersion of the UV-ozone cleaned substrate into a 1 mill molar solution of OTS in toluene starting with a 1-hour heating of the solution at 60°C. The overnight immersion was followed by rinsing with clean toluene and annealing of the clean substrates for 30 minutes at 60°C to remove all traces of solvent. The immersion, rinsing, and annealing steps were performed in a nitrogen atmosphere.

Characterization: Transistor properties were evaluated either under positive or negative gate bias to explore the majority charge carrier type and device performance (positive bias for P(NDI2OD-T2) containing devices and negative bias for PDVT-10 containing devices. Transfer curves were obtained by measuring the IDS as a function of the gate bias from -60 V to +60 V (P(NDI2OD-T2)) and from +60 V to -60 V (PDVT-10) with one point measured every 2 V, using a Keithley 2636B sourcemeter.

A xenon lamp light source connected with a monochromator, the emitted light will be introduced into the glovebox through an optical fiber and placed at a height of 5 cm from the device to provide illumination at irradiances between 0.02 to 0.2 mW cm⁻² for characterizing the photo-response of devices. We use monochromator to do the illumination test with the wavelength interval equal to 10nm. Firstly, get the dark transfer curves, waiting 100s for the recombination of the remaining electron holes in the channel, then get the illumination transfer curves, waiting 100s for the next dark transfer curves. All electrical characterization was performed in a nitrogen environment.

Supplementary Figures

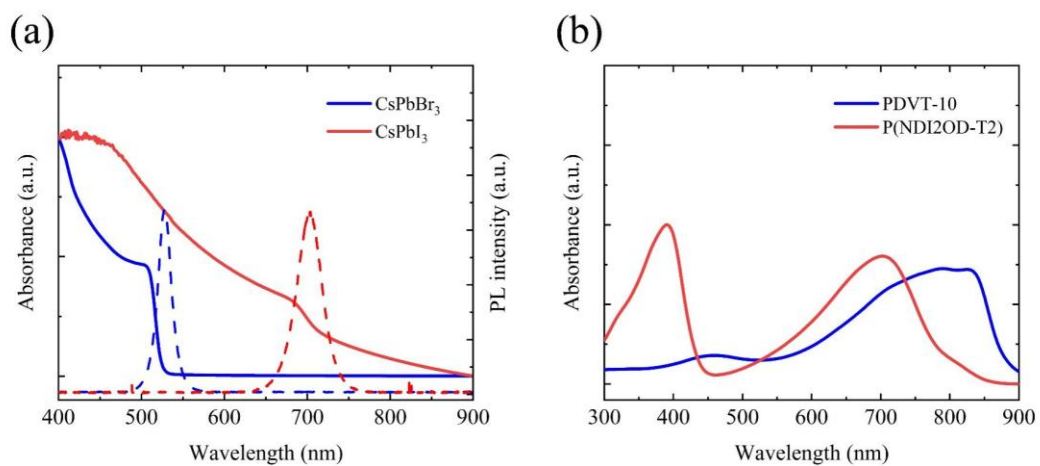


Figure S1. The absorption spectra of (a) pristine PDVT-10 and P(NDI2OD-T2) film, and (b) pristine CsPbBr₃ and CsPbI₃NCs film

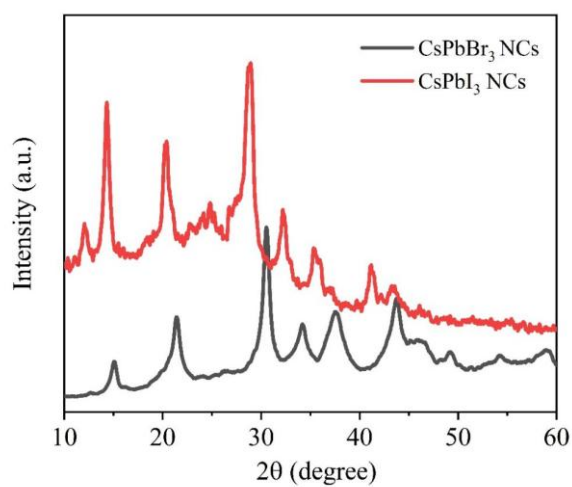


Figure S2. XRD patterns of CsPbBr₃ and CsPbI₃ NCs.

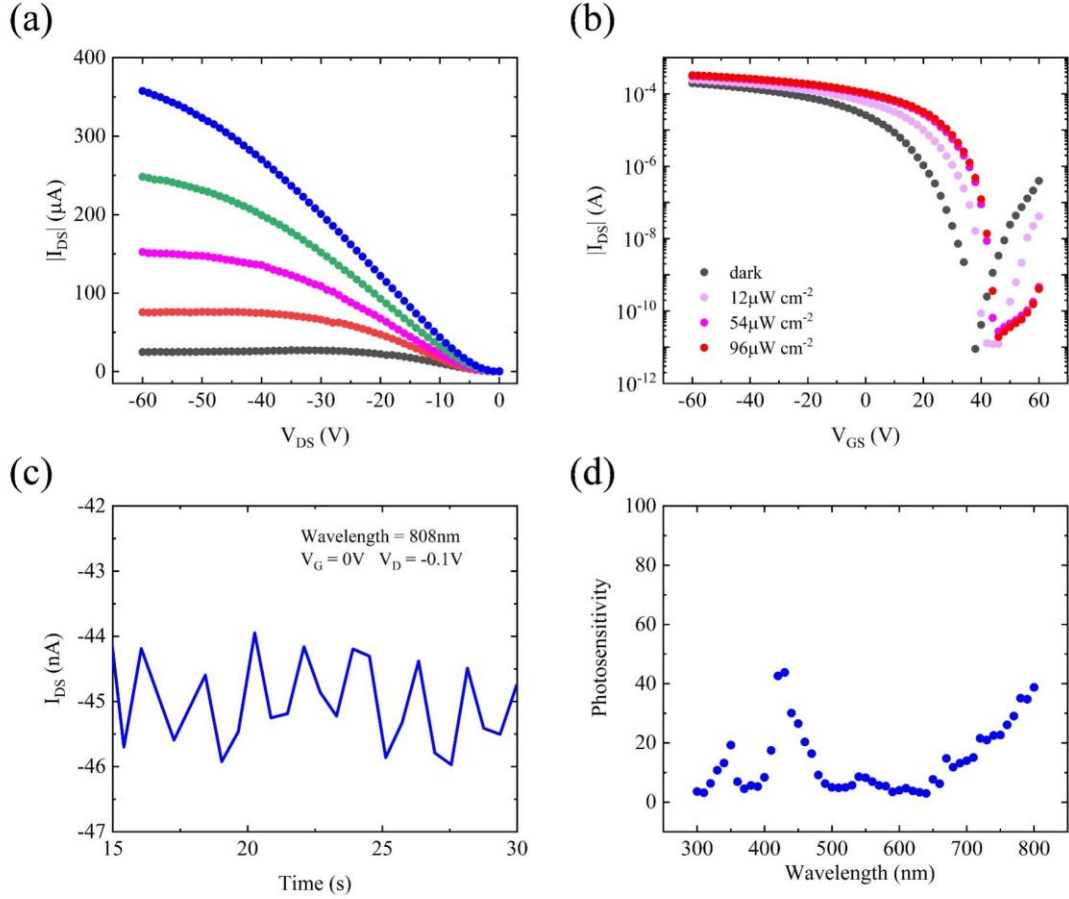


Figure S3 (a) The output curves of PDVT-10 based transistor under dark (V_{GS} swept from 0 to -40V). (b) Transfer curves of the transistor based on pure PDVT-10 under 808nm light illumination with different power intensity at a bias voltage of -20V. (c) Transient response of transistor based on PDVT-10 under 808 nm light pulse at $V_G=0$ V, $V_D=-5$ V. (d) The photoresponsivity of pure PDVT-10 measured under different wavelengths.

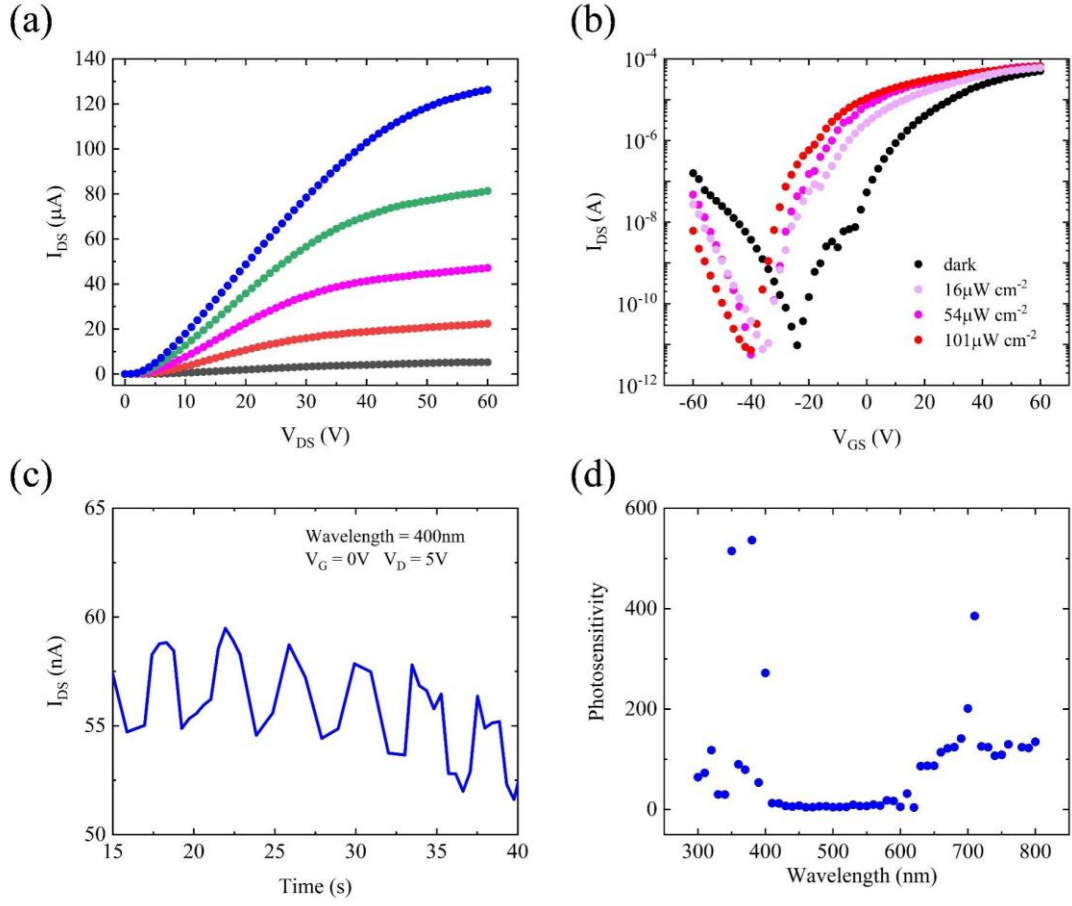


Figure S4 (a) The output curves of P(NDI2OD-T2) based transistor under dark (V_{GS} swept from 0 to 40V). (b) Transfer curves of the transistor based on pure P(NDI2OD-T2) under 400nm light illumination with different power intensity at a bias voltage of 20V. (c) Transient response of transistor based on P(NDI2OD-T2) under 400 nm light pulse at $V_G=0$ V, $V_D= 5$ V. (d) The photoresponsivity of pure P(NDI2OD-T2) measured under different wavelengths.

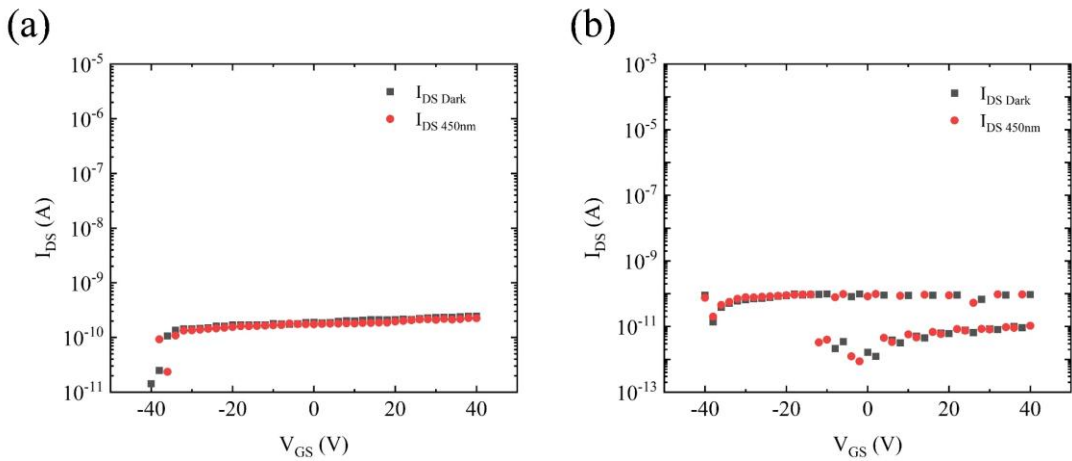


Figure S5 Transfer characteristics for (a) CsPbBr₃ and (b) CsPbI₃ NCs transistors under dark and illumination

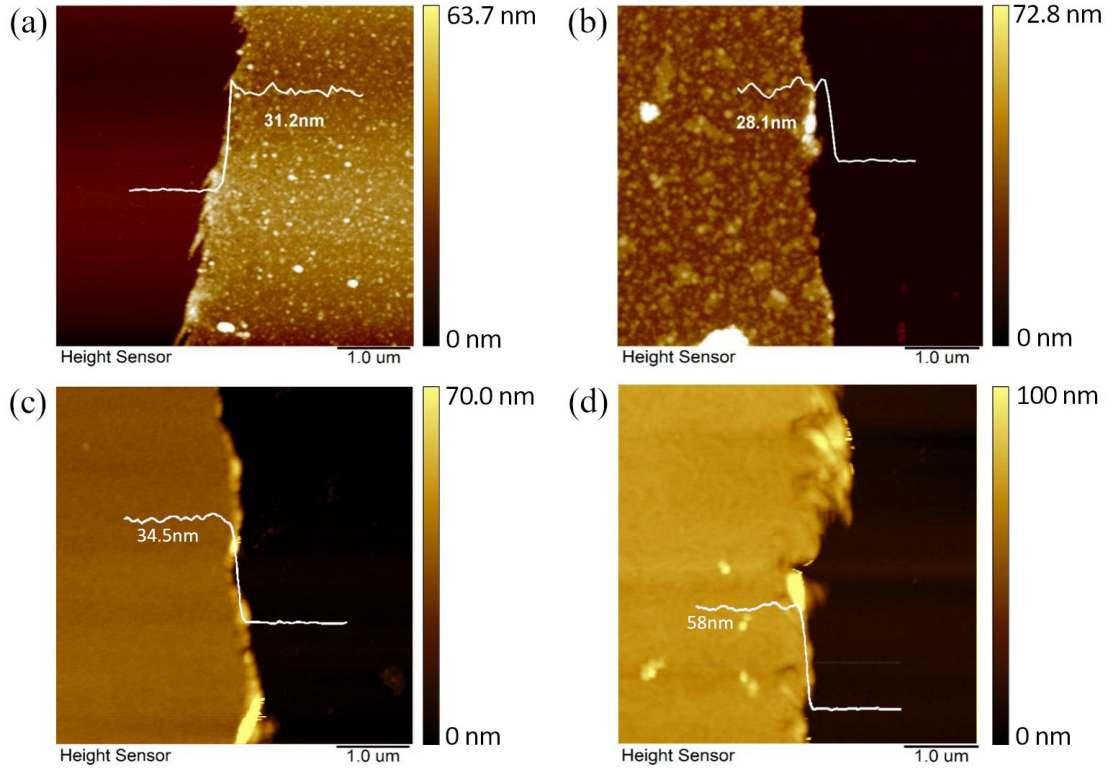


Figure S6. AFM height plot showing the thickness of (a) CsPbBr₃/PDVT-10 films, (b) CsPbI₃/P(NDI2OD-T2) films, (c) PDVT-10 films and (d) P(NDI2OD-T2) films.

The calculation formulas of Photosensitivity(P), Responsivity(R) and Detectivity(D):

$$P = \frac{I_{\text{Light}} - I_{\text{Dark}}}{I_{\text{Dark}}}$$

$$R = \frac{I_{\text{light}} - I_{\text{dark}}}{P_{\text{intensity}} WL}$$

$$D^* = R \sqrt{\frac{WL}{2eI_{\text{dark}}}}$$

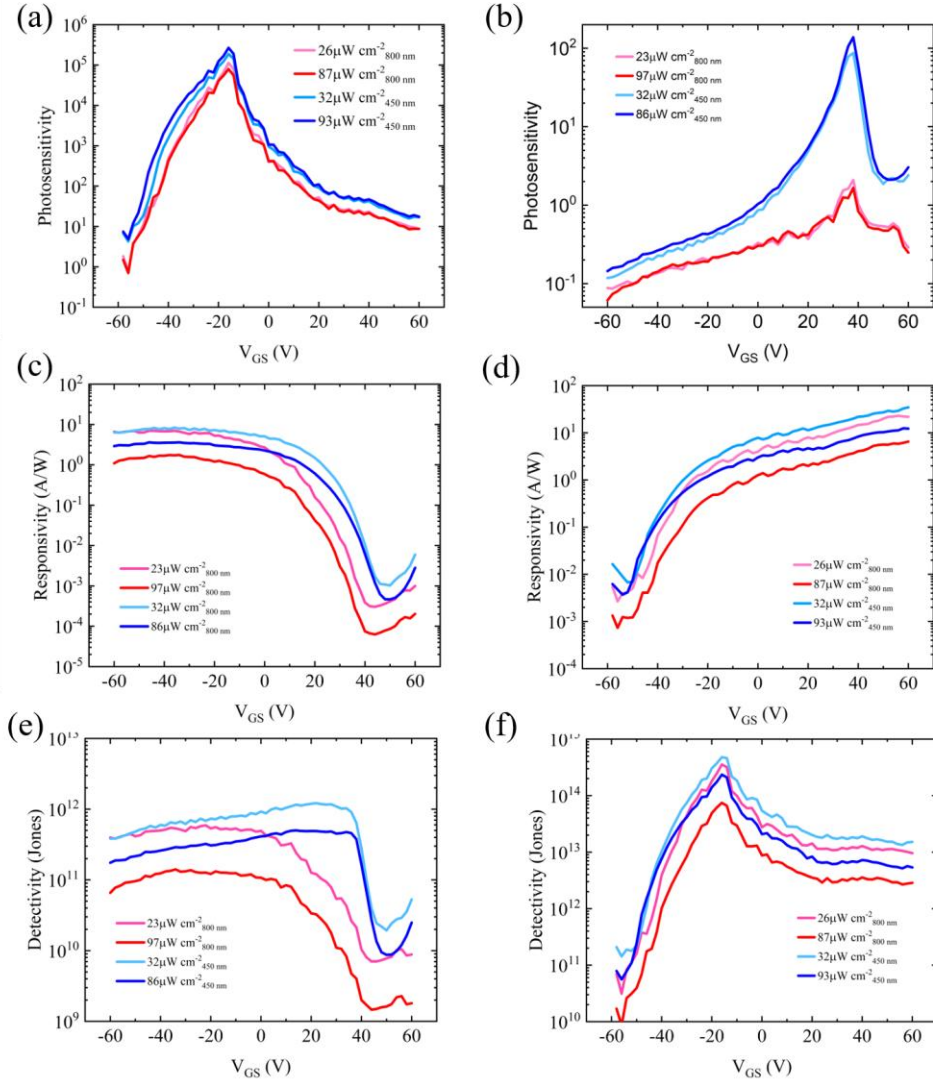


Figure S7. Photosensitivity as a function of V_{GS} of PhotoFETs based on (a) PDVT-10/CsPbBr₃ and (b) P(NDI2OD-T2)/CsPbI₃. Responsivity as a function of V_{GS} of PhotoFETs based on (c) PDVT-10/CsPbBr₃ and (d) P(NDI2OD-T2)/CsPbI₃. Detectivity as a function of V_{GS} of PhotoFETs based on (a) PDVT-10/CsPbBr₃ and (b) P(NDI2OD-T2)/CsPbI₃.

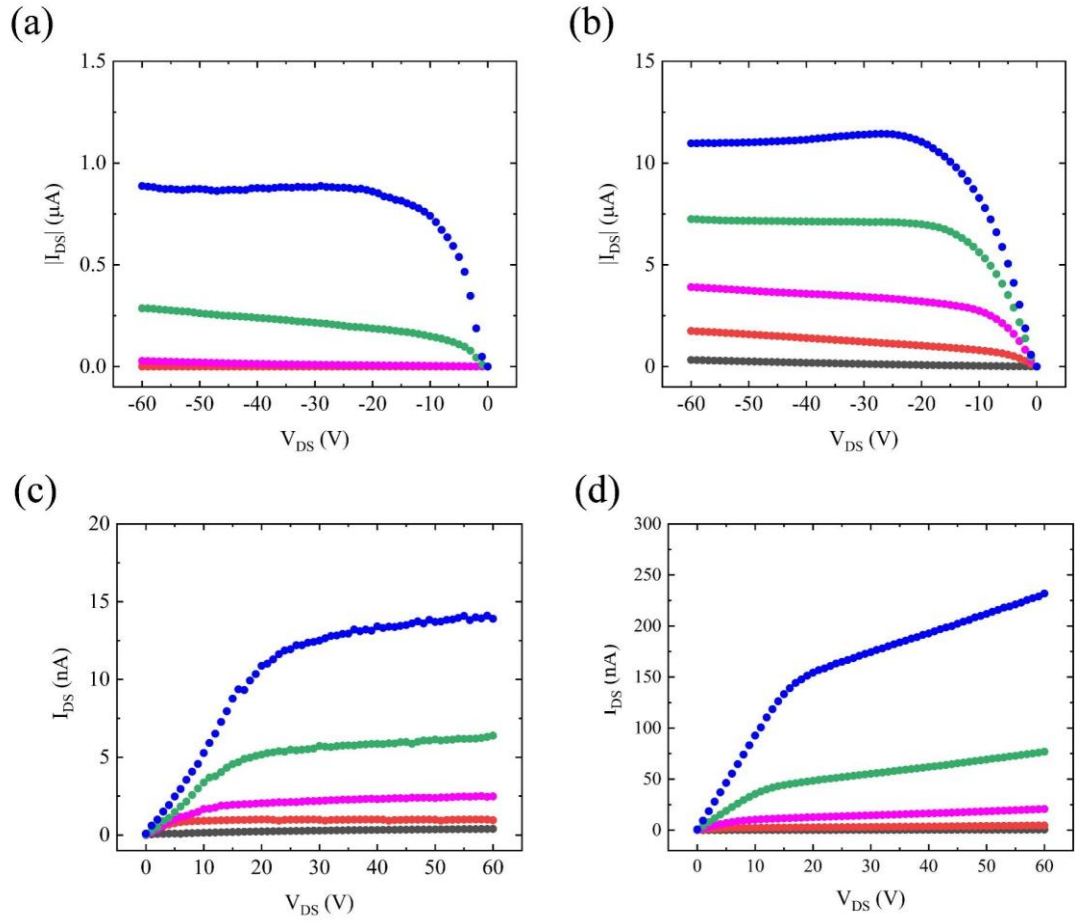


Figure S8. Output characteristics of CsPbBr₃/PDVT-10 (V_{GS} swept from 0 to 40 V) and CsPbI₃/P(NDI2OD-T2) (V_{GS} swept from 0 to -40 V) (a), (c) in dark and (b), (d) under illumination at 450 nm.

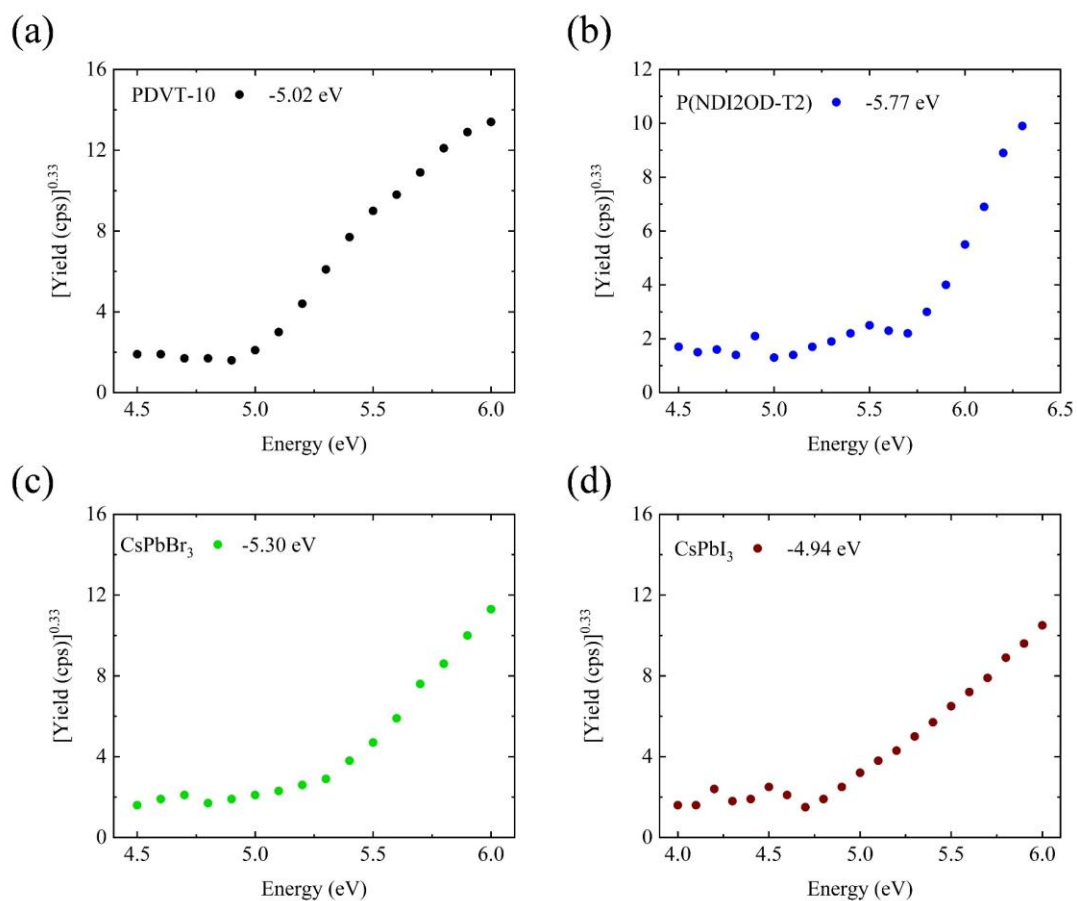


Figure S9. PESA studies carried out for HOMO of (a) PDVT-10 (b) P(NDI2OD-T2), and Ev of (c) CsPbBr₃ (d) CsPbI₃

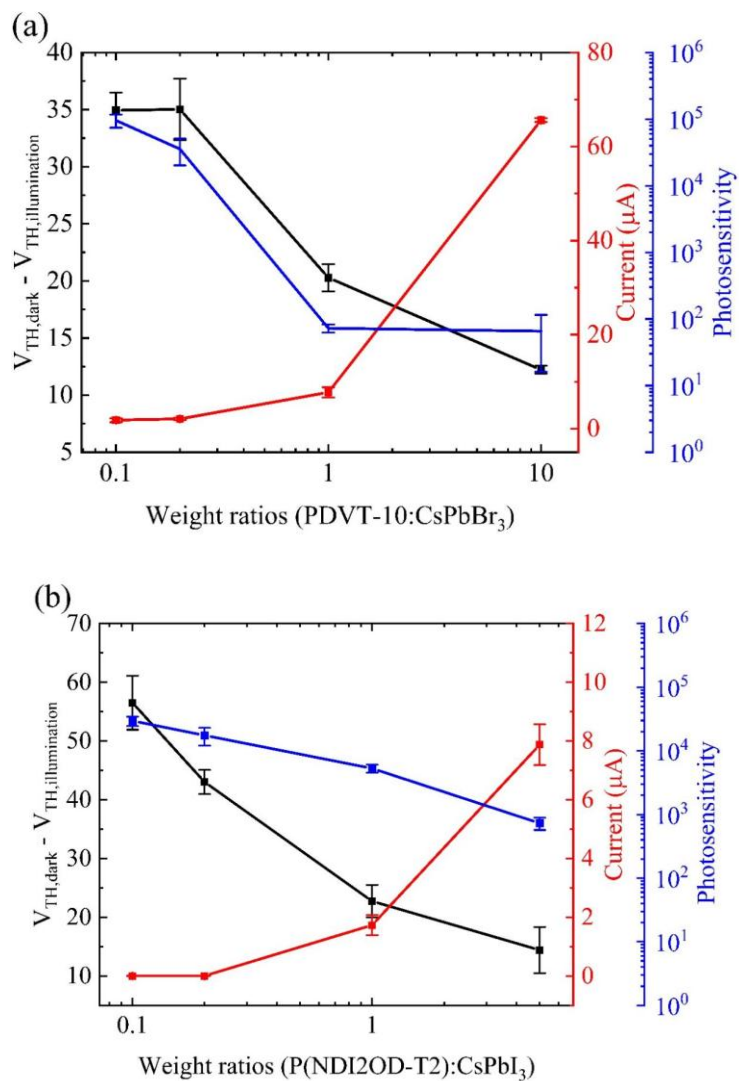


Figure S10. Threshold shift, photosensitivity and maximum current of different weight ratio.
(a) CsPbBr₃/PDVT-10 (b) CsPbI₃/P(NDI2OD-T2).

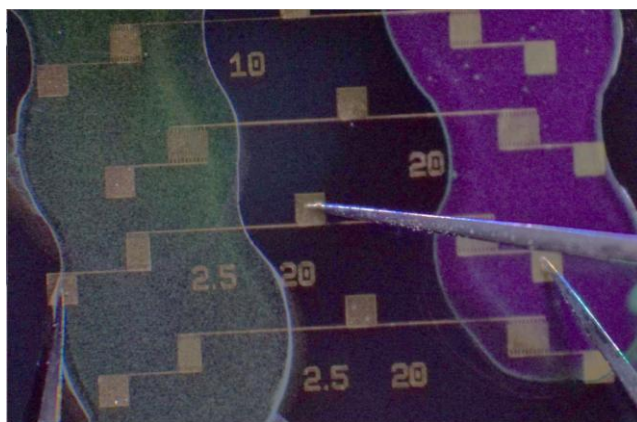


Figure S11. Optical microscope image of inverter after the two spin-coating steps.

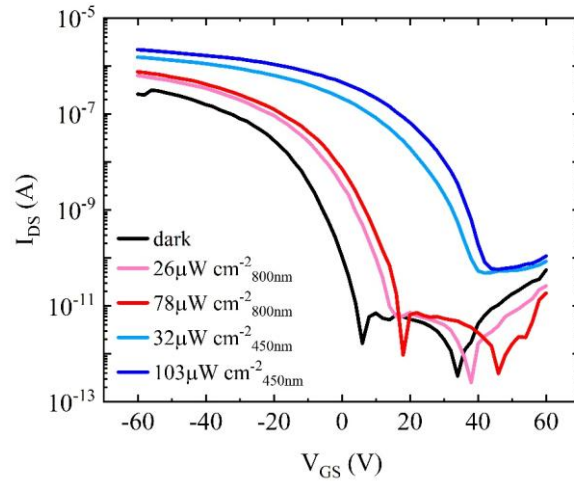


Figure S12. Transfer curves of CsPbBr₃/PDVT-10 (weight ratio=10:1) under illumination with different light densities and wavelength.

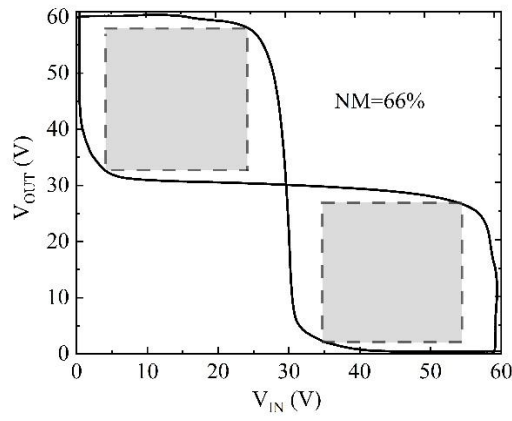


Figure S13. Butterfly inverter curves for NM calculation at $V_{DD}=60V$

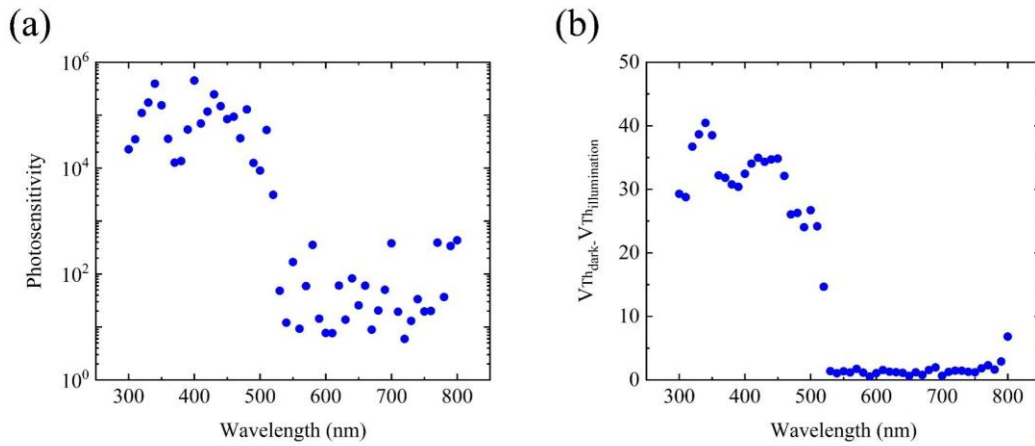


Figure S14. Performance of (a) photosensitivity and (b) threshold shift of CsPbBr₃/PDVT-10 (weight ratio=10:1) along with wavelength.

Table S1. Comparison in Device Performance of hybrid complementary inverter.

Multi-wavelength detection	N-channel material	Family	P-channel material	Family	Voltage Gain	switching voltage difference	Ref
No	Pentacene	OSC	F16CuPc	OSC	5	5/40-10/40	[1]
No	Pentacene	OSC	MoS ₂ /MAPbI ₃	TMD/PVSK	9	N/A	[2]
No	ZnO/QDs	QD	R=2M Ω	\	N/A	2.5/35	[3]
No	FACs/C16-IDTBT	OSC/PVSK	FACs/C16-IDTBT	OSC/PVSK	9	N/A	[4]
No	FACs/C8-BTBT	OSC/PVSK	FACs/C8-BTBT	OSC/PVSK	15	2/10	[4]
No	n-Si NW	NW	p-Si NW	NW	2.5	2/10	[5]
Yes	BPE-PTCDI/PDI-sol	OSC	DNTT/C10-DNTT	OSC	3	1/5	[6]
Yes	Pentacene	OSC	PTCDI-C5	OSC	N/A	6/40	[7]
Yes	P(NDI2OD-T2)/CsPbI ₃	OSC/PVSK	PDVT-10/CsPbBr ₃	OSC/PVSK	28	21/60	This work

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