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Centimeter-sized Novel Two-dimensional Organic Lead-Tin Mixed Iodide Single Crystals for Efficient Photodetector Applications

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Table S1. Crystal and Refinement Data for
(C₈H₉F₃N)₂Pb_{1-x}Sn_xI₄(x=0,0.5,1) single crystal

Empirical Formula	(C ₈ H ₉ F ₃ N) ₂ PbI ₄	(C ₈ H ₉ F ₃ N) ₂ Pb _{0.5} Sn _{0.5} I ₄	(C ₈ H ₉ F ₃ N) ₂ SnI ₄
Formula Weight/g·mol⁻¹	1067.11	1022.86	978.61
Crystal System	monoclinic	monoclinic	monoclinic
Space Group	P2 ₁ /c(14)	P2 ₁ /c(14)	P2 ₁ /c(14)
Unit Cell Dimensions	a=18.3980(8)Å	18.404(4)Å	18.3143(10)Å
	b=8.5193(3)Å	8.5313(16)Å	8.5322(4)Å
	c=8.7052(3)Å	8.6674(17)Å	8.6166(5)Å
	β=97.910(2)°	β=97.850(4)°	β=97.786(2)°
Volume/Å³	1351.45(9)	1348.1(5)	1334.03
ρ_{calculated}/g·mol⁻¹	2.622	2.520	2.436
Z	2	2	2
Index Ranges	-21≤h≤21	-21≤h≤21	-21≤h≤21
	-10≤k≤10	-9≤k≤10	-10≤k≤10
	-10≤l≤10	-8≤l≤10	-10≤l≤10
Completeness to θ = 25	100%	98.7%	100%
Data/Restraints/Parameters	2382/0/123	2345/142/17 1	2353/0/123
Goodness-of-Fit	1.062	1.086	1.021
Final R Indices [I > 2σ(I)]	R _{obs} =0.0314 ωR _{obs} =0.0654	R _{obs} =0.0361 ωR _{obs} =0.1078	R _{obs} =0.0644 ωR _{obs} =0.0630
R Indices [all data]	R _{all} =0.0439 ωR _{all} =0.0702	R _{all} =0.0418 ωR _{all} =0.1131	R _{all} =0.0363 ωR _{all} =0.0572
Largest Diff. Peak and Hole	-0.6 and 0.9 e·Å ⁻³	-0.9 and 1.2 e·Å ⁻³	-0.6 and 0.7 e·Å ⁻³
2Theta Range (Data Collection)	2.235 to 24.995	2.234 to 24.995	2.240 to 24.000
CCDC Number	2041929 [Reported]	2234210	2211886

Table S2. Fractional atomic coordinates and equivalent isotropic Displacement Parameters for $(C_8H_9F_3N)_2Pb_{1-x}Sn_xI_4$ ($x=0,0.5,1$).

$(C_8H_9F_3N)_2PbI_4$				
Atom	x/a	y/b	z/c	U [\AA^2]
Pb1	0.5	0.5	0.5	0.03854(14)
I1	0.32778(3)	0.43545(7)	0.43828(7)	0.05944(19)
I2	0.47029(3)	0.81034(6)	0.29930(6)	0.05504(18)
C1	0.2848(5)	0.9509(12)	0.4259(11)	0.071(3)
C2	0.2209(3)	0.9567(7)	0.5133(6)	0.057(2)
C3	0.2022(3)	0.8229(5)	0.5903(7)	0.065(2)
C4	0.1456(3)	0.8285(5)	0.6809(7)	0.069(3)
C5	0.1078(3)	0.9679(7)	0.6945(7)	0.062(2)
C6	0.1265(3)	1.1018(5)	0.6175(7)	0.071(3)
C7	0.1831(3)	1.0962(6)	0.5269(7)	0.075(3)
C8	0.0516(7)	0.9786(15)	0.7996(16)	0.090(3)
F1	-0.0145(5)	0.9906(13)	0.7237(12)	0.175(4)
F2	0.0439(5)	0.8528(12)	0.8795(11)	0.167(4)
F3	0.0574(6)	1.0905(13)	0.8892(13)	0.221(7)
N1	0.3503(4)	1.0236(8)	0.5193(9)	0.067(2)
H1A	0.3908	0.9912	0.4836	0.081
H1B	0.3521	0.9954	0.6182	0.081
H1C	0.3472	1.1277	0.5118	0.081
H1D	0.2731	1.0068	0.3286	0.085
H1E	0.2954	0.8426	0.4026	0.085
H3	0.2275	0.7296	0.5812	0.078
H4	0.1331	0.7389	0.7324	0.083
H6	0.1012	1.1951	0.6266	0.085
H7	0.1956	1.1857	0.4754	0.09
$(C_8H_9F_3N)_2Pb_{0.5}Sn_{0.5}I_4$				
Atom	x/a	y/b	z/c	U [\AA^2]
Pb1	0	0.5	0	0.03453(18)
Sn1	0	0.5	0	0.03453(18)
I1	0.17133(3)	0.43646(6)	0.06123(6)	0.0535(2)
I2	0.02869(3)	0.80555(5)	0.20441(5)	0.0491(2)
C1	0.2149(5)	0.5481(12)	0.5771(11)	0.066(2)
C2	0.2780(4)	0.5416(10)	0.4892(10)	0.0516(17)
C3	0.2979(5)	0.6772(11)	0.4117(11)	0.064(2)
C3B	0.4415(13)	0.519(4)	0.199(2)	0.081(5)
C4	0.3563(6)	0.6698(11)	0.3197(11)	0.068(2)
C5	0.3918(5)	0.5317(10)	0.3011(11)	0.0593(17)
C6	0.3720(5)	0.3990(11)	0.3823(12)	0.066(2)
C7	0.3168(5)	0.4050(10)	0.4734(11)	0.062(2)

C8	0.4490(7)	0.5260(16)	0.2081(15)	0.091(2)
F1	0.5170(5)	0.5189(16)	0.2861(15)	0.109(3)
F1B	0.5034(15)	0.438(4)	0.237(4)	0.089(7)
F2	0.4586(6)	0.6562(13)	0.1270(13)	0.109(3)
F2B	0.4204(17)	0.585(4)	0.062(3)	0.090(6)
F3	0.4162(14)	0.463(4)	0.055(3)	0.085(6)
F3B	0.4500(7)	0.3961(13)	0.1202(15)	0.114(3)
N1	0.1477(4)	0.4747(8)	0.4858(9)	0.0605(18)
H1A	0.15	0.4842	0.3843	0.073
H1B	0.1077	0.5229	0.5089	0.073
H1C	0.1458	0.3736	0.5103	0.073
H1D	0.2045	0.6565	0.5999	0.079
H1E	0.2269	0.4932	0.6752	0.079
H3	0.273	0.771	0.4206	0.076
H4	0.3704	0.7601	0.2716	0.082
H6	0.3969	0.3052	0.3739	0.079
H7	0.3051	0.3152	0.5258	0.075
(C₈H₉F₃N)₂SnI₄				
Atom	x/a	y/b	z/c	U [Å²]
Sn1	0.5	1	0.5	0.0373(2)
I1	0.52773(3)	0.80061(5)	0.20851(6)	0.05400(18)
I2	0.32929(3)	0.93706(6)	0.43917(6)	0.05812(19)
C1	0.2865(5)	0.4514(10)	0.4174(10)	0.069(2)
C2	0.2219(2)	0.4578(6)	0.5084(6)	0.055(2)
C3	0.2030(3)	0.3238(5)	0.5856(6)	0.059(2)
C4	0.1464(3)	0.3294(5)	0.6780(6)	0.067(2)
C5	0.1089(3)	0.4689(7)	0.6933(6)	0.063(2)
C6	0.1278(3)	0.6029(5)	0.6162(7)	0.076(3)
C7	0.1844(3)	0.5973(5)	0.5237(6)	0.071(3)
C8	0.0523(6)	0.4800(14)	0.7980(15)	0.091(3)
F1	-0.0130(4)	0.4953(12)	0.7251(10)	0.181(4)
F2	0.0596(5)	0.5883(12)	0.8938(12)	0.237(7)
F3	0.0441(5)	0.3549(10)	0.8773(9)	0.164(3)
N1	0.3529(3)	0.5230(7)	0.5119(8)	0.069(2)
H1A	0.3933	0.4938	0.4725	0.082
H1B	0.3558	0.4907	0.6108	0.082
H1C	0.349	0.6269	0.5085	0.082
H1D	0.2969	0.3433	0.3931	0.083
H1E	0.2748	0.5079	0.3195	0.083
H3	0.2281	0.2305	0.5753	0.071
H4	0.1337	0.2397	0.7297	0.08
H6	0.1027	0.6962	0.6264	0.091

H7	0.1971	0.687	0.4721	0.085
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Table S3. Bond lengths of $(C_8H_9F_3N)_2Pb_{1-x}Sn_xI_4$ ($x=0,0.5,1$).

$(C_8H_9F_3N)_2PbI_4$		$(C_8H_9F_3N)_2SnI_4$	
Atoms	d	Atoms	d
Pb1—I2	3.1747(5)	Sn1—I1 ⁱ	3.1315(5)
Pb1—I2 ⁱ	3.1765(5)	Sn1—I1 ⁱⁱ	3.1332(5)
Pb1—I2 ⁱⁱ	3.1747(5)	Sn1—I1	3.1315(5)
Pb1—I2 ⁱⁱⁱ	3.1765(5)	Sn1—I1 ⁱⁱⁱ	3.1332(5)
Pb1—I1 ⁱⁱ	3.1878(6)	Sn1—I2	3.1454(6)
Pb1—I1	3.1878(6)	Sn1—I2 ⁱ	3.1454(6)
I2—Pb1 ^{iv}	3.1765(5)	I1—Sn1 ^{iv}	3.1332(5)
N1—C1	1.4924(114)	N1—C1	1.4989(101)
C1—C2	1.4865(115)	F3—C8	1.2871(149)
F3—C8	1.2271(173)	C2—C7	1.3893(67)
F1—C8	1.3057(152)	C2—C3	1.3899(70)
F2—C8	1.2960(167)	C2—C1	1.5063(105)
C6—C5	1.3901(79)	C7—C6	1.3914(83)
C6—C7	1.3909(87)	C6—C5	1.3895(77)
C5—C4	1.3902(75)	C5—C4	1.3893(75)
C5—C8	1.4747(155)	C5—C8	1.4665(139)
C4—C3	1.3909(87)	C4—C3	1.3908(81)
C3—C2	1.3894(77)	F1—C8	1.2800(129)
C2—C7	1.3902(79)	F2—C8	1.2342(160)
$(C_8H_9F_3N)_2Pb_{0.5}Sn_{0.5}I_4$			
Atoms	d	Atoms	d
Pb1 Sn1—I2 ⁱ	3.1555(6)	C4—C3	1.425(15)
Pb1 Sn1—I2 ⁱⁱ	3.1566(6)	C5—C6	1.406(12)
Pb1 Sn1—I2	3.1556(6)	C5—C3B	1.359(15)
Pb1 Sn1—I2 ⁱⁱⁱ	3.1566(6)	C5—C8	1.411(12)
Pb1 Sn1—I1	3.1719(8)	C7—C6	1.371(13)
Pb1 Sn1—I3 ⁱ	3.1720(8)	Pb01 Sn01—I002 ⁱⁱ	3.1566(6)
I2—Pb1 Sn1 ^{iv}	3.1566(6)	Pb01 Sn01—I002 ⁱ	3.1555(6)
I2—Pb1 Sn1 ^{iv}	3.1566(6)	Pb01 Sn01—I002 ⁱⁱⁱ	3.1566(6)
I2—Pb1 Sn1	3.1556(6)	Pb01 Sn01—I003 ⁱ	3.1720(8)
I1—Pb1 Sn1	3.1719(8)	F1B—C3B	1.329(15)
N1—C1	1.509(11)	F2B—C3B	1.333(15)
C2—C7	1.383(12)	C3B—F4B	1.363(16)
C2—C3	1.412(13)	C3A—F4A	1.346(12)
C2—C1	1.474(12)	C3A—F2A	1.340(12)
C4—C5	1.368(14)	C3A—F1A	1.339(12)
(i) -x, 1-y, -z; (ii) x, 1.5-y, -0.5+z; (iii) -x, -0.5+y, 0.5-z; (iv) -x, 0.5+y, 0.5-z.			

Table S4. Bond angles of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$).

$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$		$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$	
Atoms	Angle	Atoms	Angle
I2—Pb1—I2 ⁱⁱ	180.000	I1 ⁱ —Sn1—I1	180.000
I2 ⁱⁱ —Pb1—I2 ⁱ	89.528(13)	I1—Sn1—I1 ⁱⁱ	90.923(12)
I2—Pb1—I2 ⁱ	90.472(13)	I1 ⁱ —Sn1—I1 ⁱⁱ	89.077(12)
I2—Pb1—I2 ⁱⁱⁱ	89.528(13)	I1 ⁱ —Sn1—I1 ⁱⁱⁱ	90.923(12)
I2 ⁱⁱ —Pb1—I2 ⁱⁱⁱ	90.472(13)	I1—Sn1—I1 ⁱⁱⁱ	89.077(12)
I2 ⁱⁱⁱ —Pb1—I2 ⁱ	180.000	I1 ⁱⁱⁱ —Sn1—I1 ⁱⁱ	180.000(12)
I2 ⁱⁱⁱ —Pb1—I1	86.948(15)	I1 ⁱⁱ —Sn1—I2	87.937(14)
I2—Pb1—I1	87.653(15)	I1—Sn1—I2	92.242(14)
I2 ⁱ —Pb1—I1 ⁱⁱ	86.948(15)	I1 ⁱ —Sn1—I2	87.758(14)
I2 ⁱⁱ —Pb1—I1 ⁱⁱ	87.653(15)	I1 ⁱⁱⁱ —Sn1—I2 ⁱ	87.937(14)
I2 ⁱⁱ —Pb1—I1	92.347(15)	I1 ⁱⁱⁱ —Sn1—I2	92.063(14)
I2 ⁱⁱⁱ —Pb1—I1 ⁱⁱ	93.052(15)	I1 ⁱⁱ —Sn1—I2 ⁱ	92.063(14)
I2—Pb1—I1 ⁱⁱ	92.347(15)	I1 ⁱ —Sn1—I2 ⁱ	92.242(14)
I2 ⁱ —Pb1—I1	93.052(15)	I1—Sn1—I2 ⁱ	87.758(14)
I1 ⁱⁱ —Pb1—I1	180.000	I2 ⁱ —Sn1—I2	180.000
Pb1—I2—Pb1 ^{iv}	147.035(18)	Sn1—I1—Sn1 ^{iv}	150.849(16)
C2—C1—N1	110.094(689)	C7—C2—C3	120.062(443)
C5—C6—C7	119.970(503)	C7—C2—C1	120.909(491)
C6—C5—C8	119.314(691)	C3—C2—C1	118.908(494)
C4—C5—C6	120.035(496)	C6—C7—C2	120.000(445)
C4—C5—C8	120.486(681)	C7—C6—C5	119.922(482)
C3—C4—C5	119.961(482)	C6—C5—C8	118.921(642)
C4—C3—C2	120.022(489)	C4—C5—C6	120.079(493)
C3—C2—C1	119.041(578)	C4—C5—C8	120.889(635)
C7—C2—C1	120.807(586)	C3—C4—C5	120.007(470)
C7—C2—C3	120.017(506)	C4—C3—C2	119.932(447)
C2—C7—C6	119.995(496)	N1—C1—C2	109.821(631)
F3—C8—F1	104.732(1186)	F3—C8—C5	114.586(947)
F3—C8—F2	107.920(1162)	F1—C8—F3	100.175(1009)
F3—C8—C5	115.805(1125)	F1—C8—C5	113.327(878)
F1—C8—C5	112.001(1009)	F2—C8—F3	106.003(1055)
F2—C8—F1	99.982(1095)	F2—C8—C5	115.992(1018)
F2—C8—C5	114.809(1047)	F2—C8—F1	105.143(1061)
(i) 1-x, 2-y, 1-z; (ii) 1-x, 0.5+y, 0.5-z; (iii) x, 1.5-y, 0.5+z; (iv) 1-x, -0.5+y, 0.5-z.			
$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$			
Atoms	Angle	Atoms	Angle
I2 ⁱ —Pb1 Sn1—I2	180.000	C4—C5—C6	118.2(8)
I2—Pb1 Sn1—I2 ⁱⁱⁱ	90.696(14)	C4—C5—C8	120.2(9)

$I2^i-Pb1 Sn1-I2^{iii}$	89.304(15)	C6—C5—C8	121.5(9)
$I2^i-Pb1 Sn1-I2^{ii}$	90.696(15)	C3B—C5—C4	121.4(16)
$I2-Pb1 Sn1-I2^{ii}$	89.304(14)	C3B—C5—C6	120.3(17)
$I2^{ii}-Pb1 Sn1-I2^{iii}$	180.000(19)	C6—C7—C2	121.4(8)
$I2^{ii}-Pb1 Sn1-I1^i$	92.696(15)	C2—C3—C4	119.6(9)
$I2^i-Pb1 Sn1-I1^i$	87.761(14)	C7—C6—C5	121.4(8)
$I2^{iii}-Pb1 Sn1-I1$	92.696(15)	C5—C3B—F3	117(2)
$I2-Pb1 Sn1-I1$	87.760(14)	F2B—C3B—C5	114.1(18)
$I2-Pb1 Sn1-I1^i$	92.240(14)	F2B—C3B—F1B	125.(2)
$I2^{ii}-Pb1 Sn1-I1$	87.304(15)	F2B—C3B—F3	45.(2)
$I2^i-Pb1 Sn1-I1$	92.238(14)	F1B—C3B—C5	120.9(19)
$I2^{iii}-Pb1 Sn1-I1^i$	87.304(15)	F1B—C3B—F3	102(3)
$I1-Pb1 Sn1-I1^i$	180.000	F3B—C8—C5	114.9(10)
$Pb1 Sn1-I2-Pb1 Sn1^{iv}$	148.884(18)	F1—C8—C5	115.5(11)
C2—C1—N1	111.1(7)	F1—C8—F3B	99.4(11)
C7—C2—C3	118.2(8)	F2—C8—C5	115.7(11)
C7—C2—C1	122.5(8)	F2—C8—F3B	111.9(12)
C3—C2—C1	119.3(9)	F2—C8—F1	96.9(11)
C5—C4—C3	120.9(9)		
(i) 1-x, 2-y, 1-z; (ii) 1-x, 0.5+y, 0.5-z; (iii) x, 1.5-y, 0.5+z; (iv) 1-x, -0.5+y, 0.5-z.			

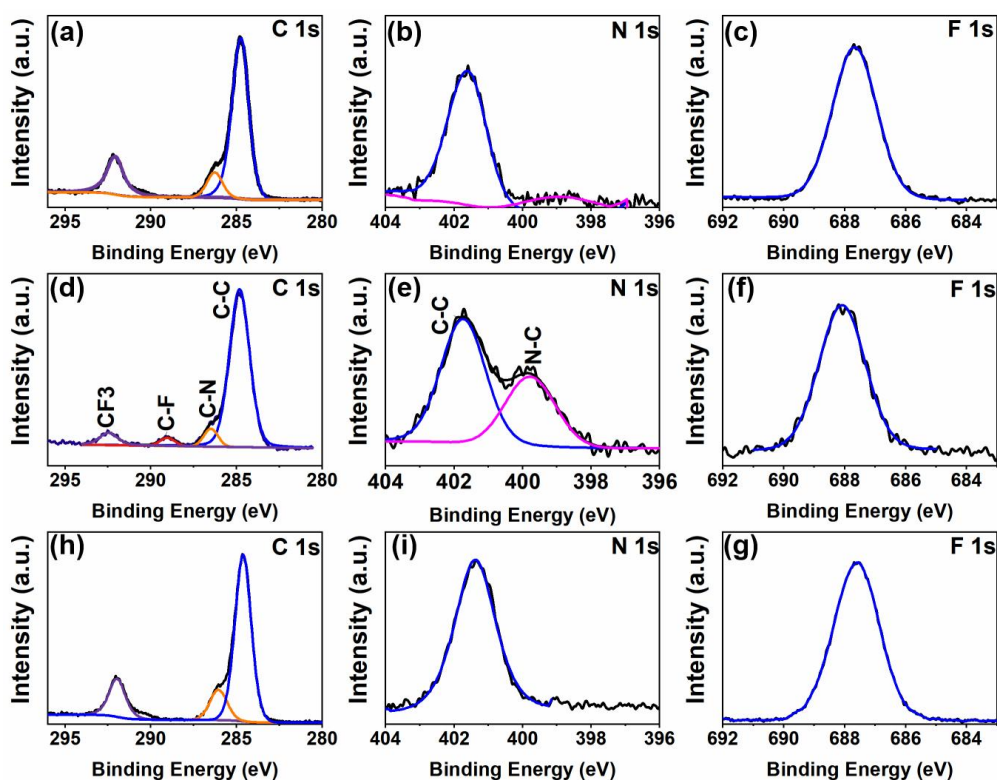


Fig. S1. High-resolution XPS spectra of $(C_8H_9F_3N)_2Pb_{1-x}Sn_xI_4$ ($x=0,0.5,1$) single crystal. (a)-(c) $(C_8H_9F_3N)_2PbI_4$, (d-f) $(C_8H_9F_3N)_2Pb_{0.5}Sn_{0.5}I_4$, and (h-g) $(C_8H_9F_3N)_2SnI_4$.

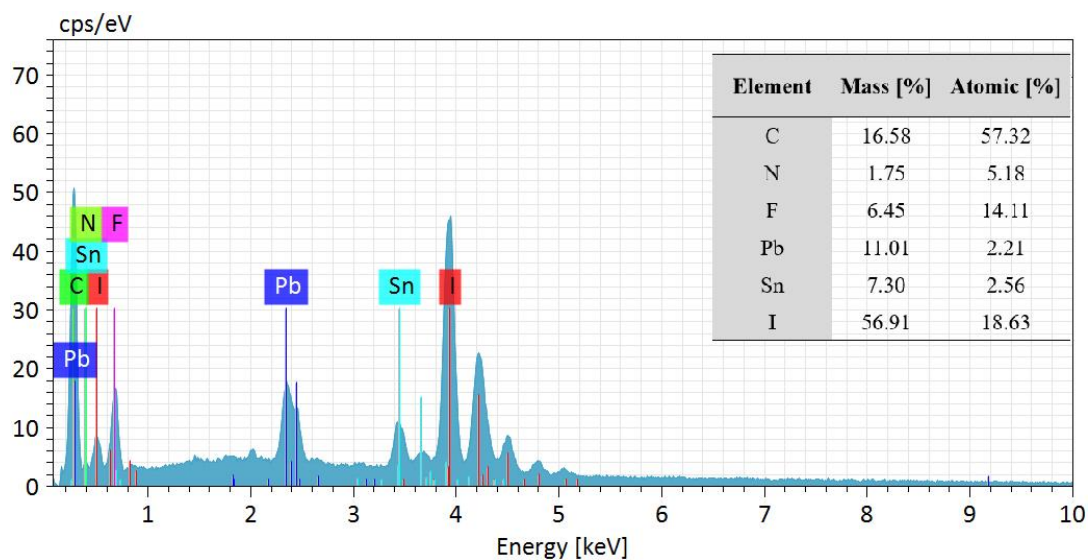


Fig. S2. SEM-EDS results of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$ single crystal.

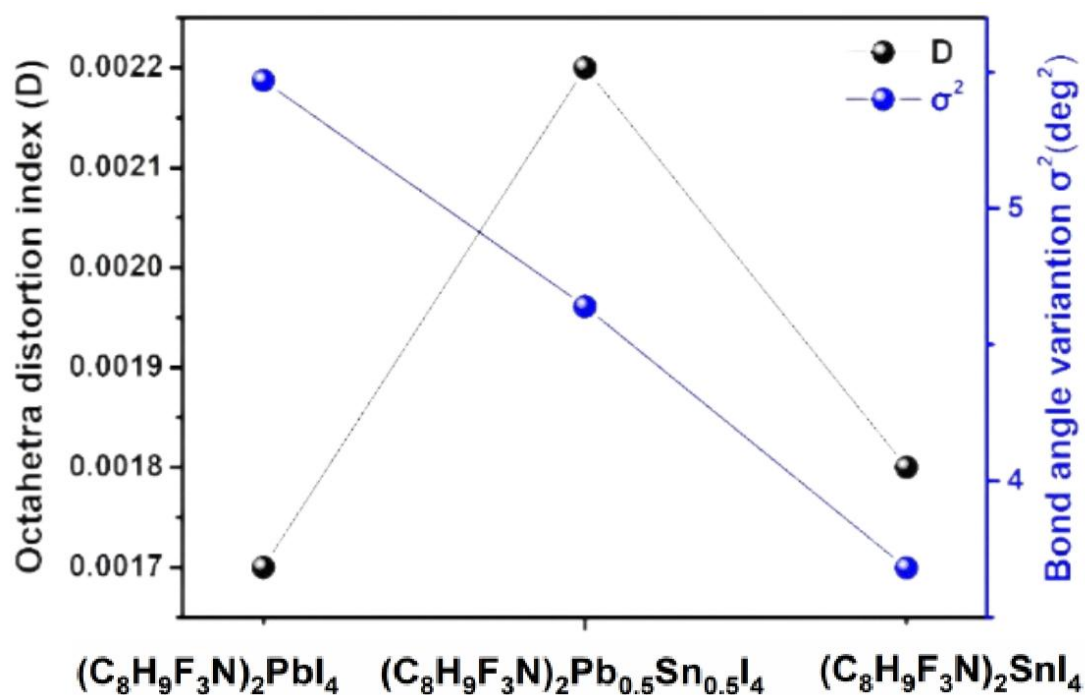


Fig. S3. Distortion Index (D), Bond Angle Variance (σ^2), and bandgap of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$).

Table S5. Distortion index (D), bond angle variance (σ^2), and bandgap of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$).

	$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$	$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$	$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$
Equatorial I-M-I angle (deg)	89.528	90.696	90.923
	90.472	89.304	89.077
	86.948	87.761	87.937
Axial I-M-I angle (deg)	87.653	87.305	92.242
	92.347	92.239	87.758
	93.052	92.695	92.063
M-I-M angle (deg)	147.035	150.849	148.884
D	0.0017	0.0022	0.0018
σ^2	5.47	4.64	3.68
Band gap (eV)	2.39	1.96	2.0

Table S6. Water contact angle of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$).

Perovskite	Water contact angle (deg)
$\text{Cs}_2\text{AgBiBr}_6/\text{SnO}_2/\text{ZnO}$ [1]	15
$\text{Cs}_2\text{AgBiBr}_6/\text{ZnO}$ [1]	48
3D MAPbI ₃ [2]	38.88
FAPbI ₃ [3]	46.9
(iBA) ₂ PbI ₄ [4]	46.2
iBA(DMPDA) _{0.5} PbI ₄ [4]	50.7
$\text{Cs}_2\text{AgBiBr}_6$ [5]	50.2
(FAPbI ₃) _{0.95} (MAPbBr ₃) _{0.05} [6]	54.6
$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$ (This work)	58.25
$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$ (This work)	60.25
$(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$ (This work)	50.00

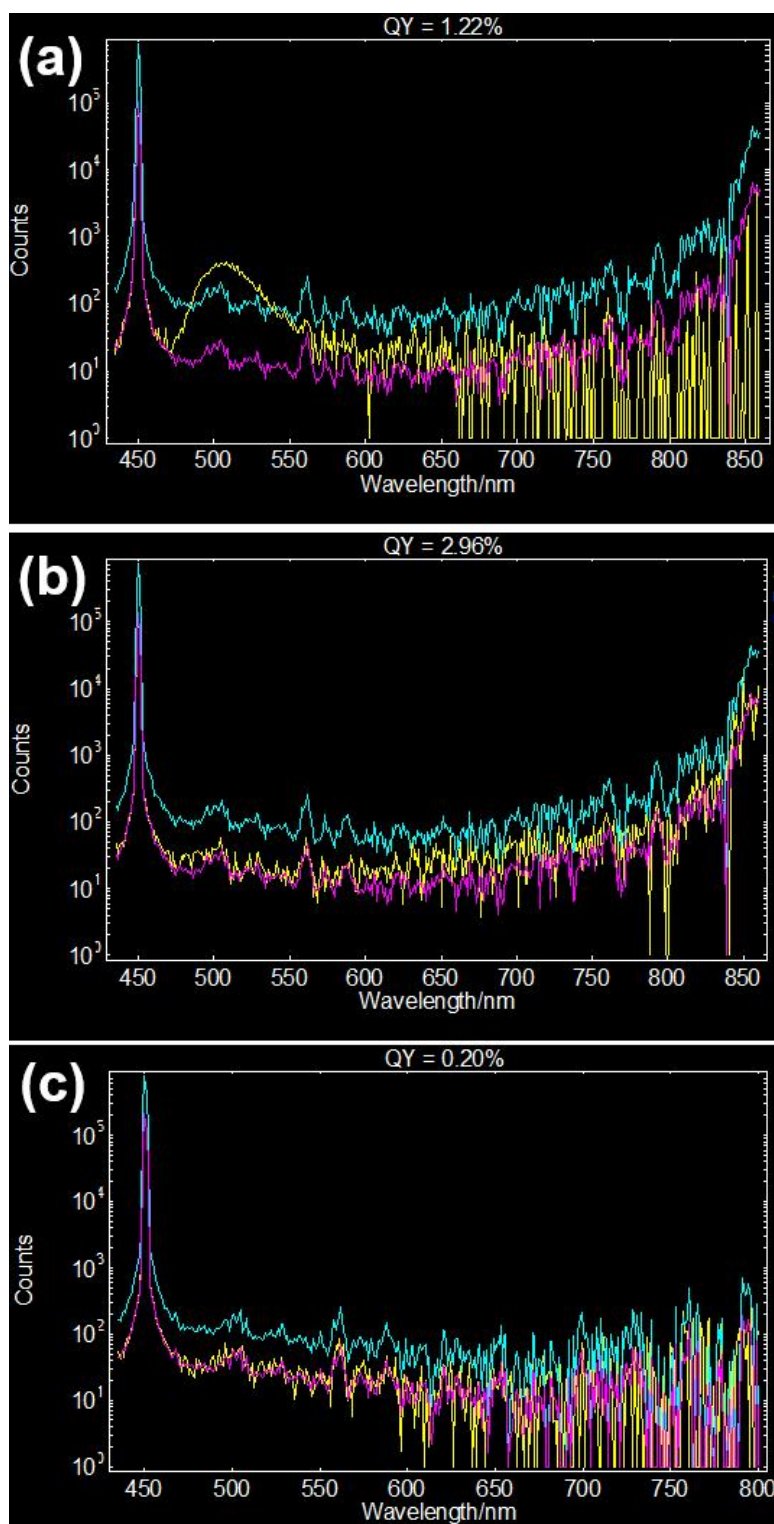


Fig. S4. PLQY results of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$). (a) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$, (b) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$, and (c) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$.

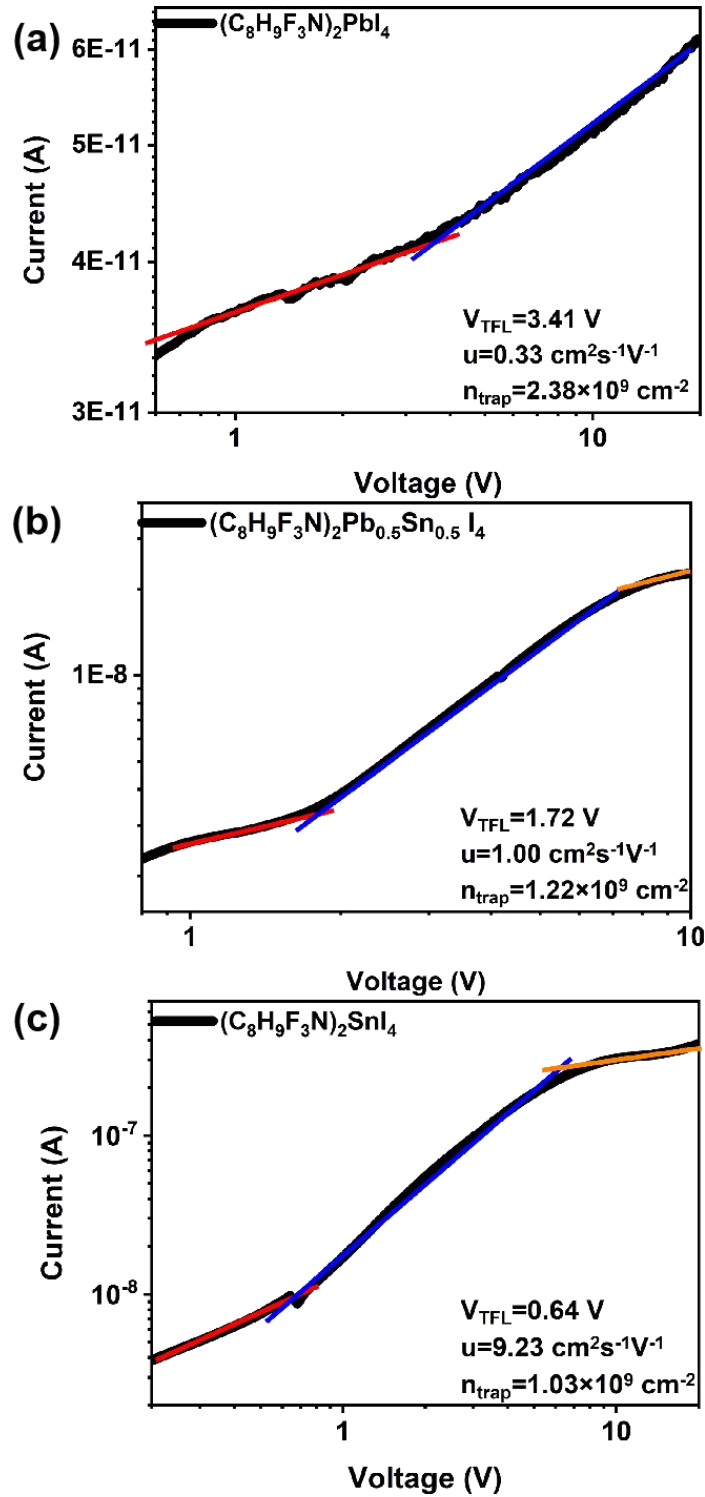


Fig. S5. Current-voltage curves and trap density at 298 K. Characteristic I - V curves with two different regimes for (a) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$ and three different regimes for (b) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$ and (c) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$. A linear Ohmic regime ($I \propto V$, red line) is followed by the trap-filled regime (TFL), marked by a steep increase in current ($I \propto V^n$ ($n > 3$)). The orange line denotes the trap-free Child's regime ($I \propto V^2$).

Table S7. The performances of Sn-containing perovskite-based photodetectors.

Materials	Light intensity ($\mu\text{W cm}^{-2}$)	On/off current ratio	R (A W^{-1})	D* (Jones)
PEA ₂ SnI ₄ film (30% SnF ₂)[7]	2000	/	207.5	2.53×10^{13}
FASnI ₃ Film[8]	0.064	/	10^5	1.9×10^{12}
TBASnCl ₃ QDs[9]	360	67	0.0148	5.04×10^{10}
TBASnCl ₃ (0.03 M SnF ₂) QDs[9]	360	238	0.0097	7.67×10^{10}
MAPb _{0.5} Sn _{0.5} I ₃ thin films[10]	614.3	/	0.0016	3.08×10^{10}
(PEA) ₂ SnI ₄ /MoS ₂ [11]	36 pw	10^2	0.121	8.09×10^9
(rGO/PEDOT:PSS)/(PEA) ₂ SnI ₄ [12]	56.9	/	16	1.92×10^{11}
TiO ₂ /CsSnI ₃ /P3HT[13]	/	/	0.257	1.5×10^{11}
CsSnI ₃ [14]	/	1.08	0.054	3.85×10^5
CH ₃ NH ₃ Pb _{0.7} Sn _{0.3} I ₃ [15]	/	/	0.39	7×10^{12}
(PEA) ₂ SnI ₄ microsheets[16]	195.8	10	3.29×10^3	2.06×10^{11}
(C ₈ H ₉ F ₃ N) ₂ PbI ₄ (This work)	7000	10^3	0.0138	8.7×10^{10}
(C ₈ H ₉ F ₃ N) ₂ Pb _{0.5} Sn _{0.5} I ₄ (This work)	7000	48	0.6681	8.6×10^{10}
(C ₈ H ₉ F ₃ N) ₂ SnI ₄ (This work)	7000	18	0.0323	4.7×10^9

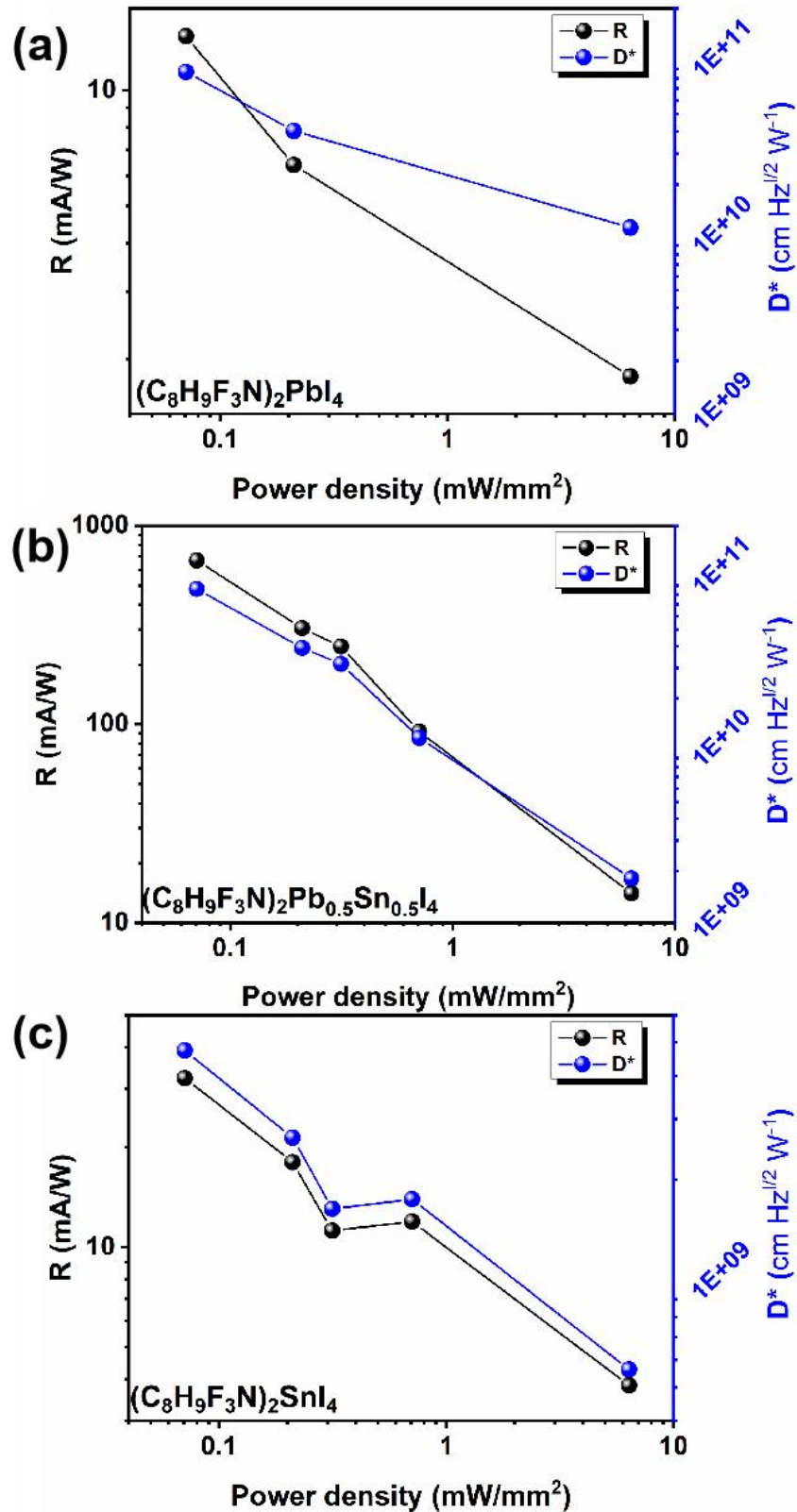


Fig. S6. Relationship of responsivity (R) and detectivity (D^*) with light power density at 10 V. (a) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$, (b) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$, and (c) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$.

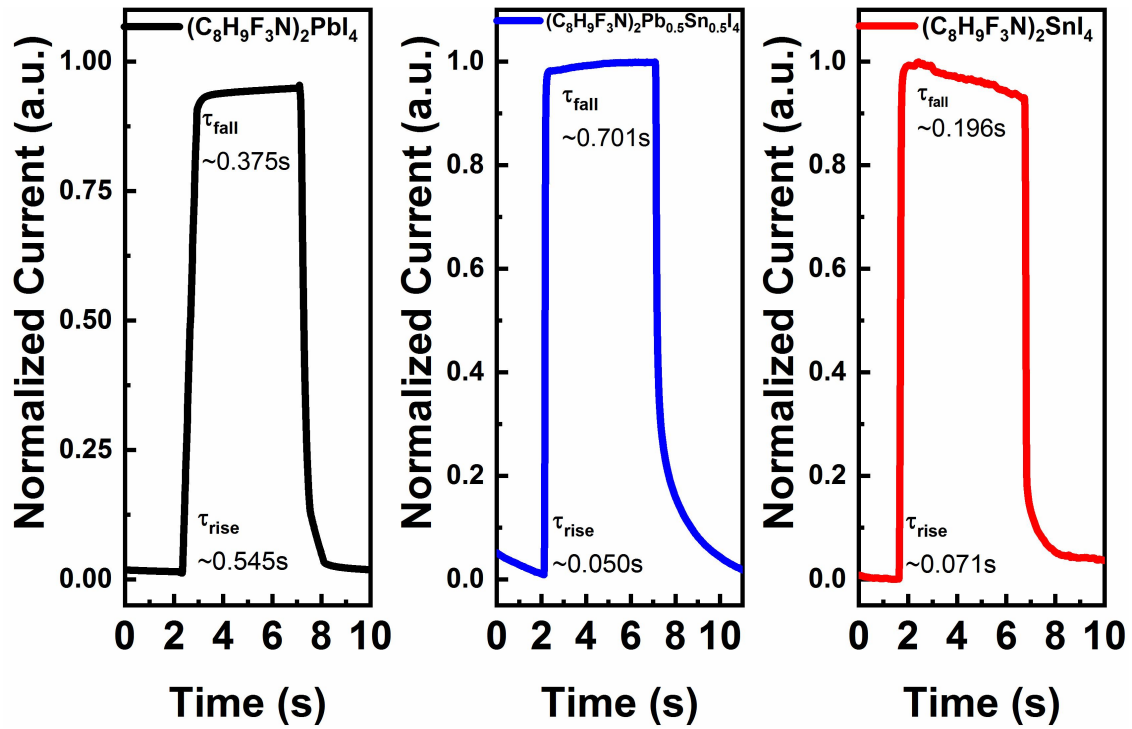


Fig. S7. Temporal response of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$) single crystal-based photodetectors at the light intensity of 20 mWcm^{-2} under a bias of 5V. (a) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$, (b) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$, and (c) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$.

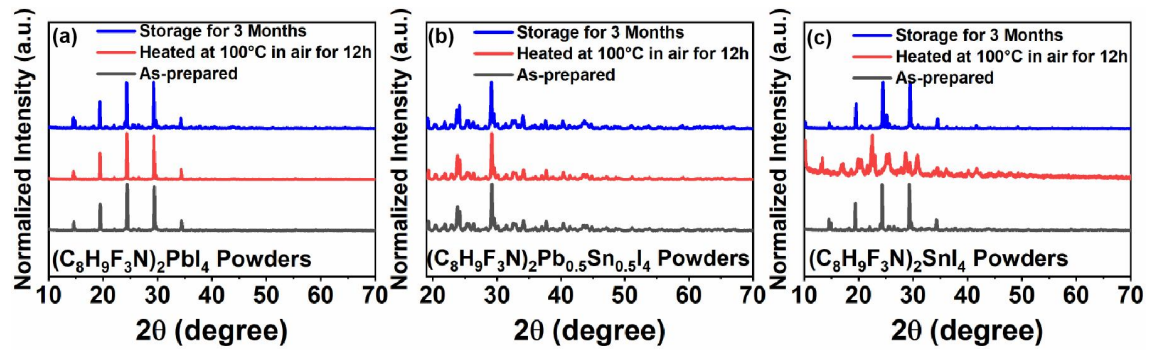


Fig. S8. Stability test. (a)-(c) PXR patterns of $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$) powders that stored in ambient environment ($T=24\text{-}30^\circ\text{C}$, $\text{RH}=45\text{-}65\%$) for above 3 months.

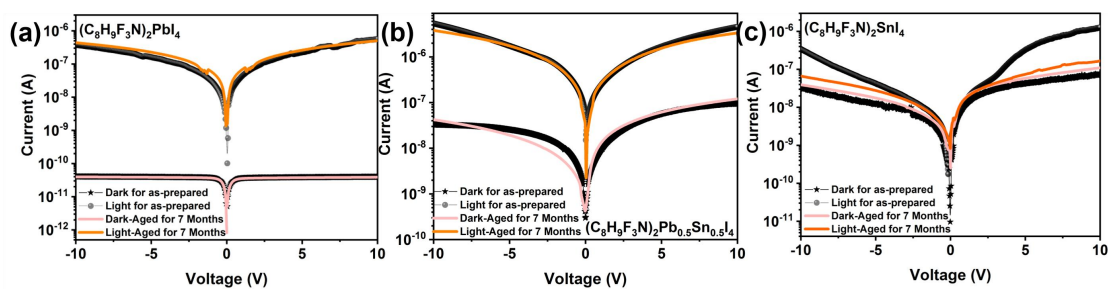


Fig. S9. Stability test. I-V curves for (a) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{PbI}_4$, (b) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{0.5}\text{Sn}_{0.5}\text{I}_4$, and (c) $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{SnI}_4$ with the device architecture of Au/BCP/ C_{60} / $(\text{C}_8\text{H}_9\text{F}_3\text{N})_2\text{Pb}_{1-x}\text{Sn}_x\text{I}_4$ ($x=0,0.5,1$) single crystal/ C_{60} /BCP/Au under the illumination of 405 nm laser ($P=6.37$ mW/mm^2).

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