Low dark current and Type-II band Alignment in Double Perovskite Single Crystal $Cs_2AgBiBr_6$ / $Cs_3Bi_2I_9$ Nanocrystals Heterojunction Enables High-Performance Photodetection

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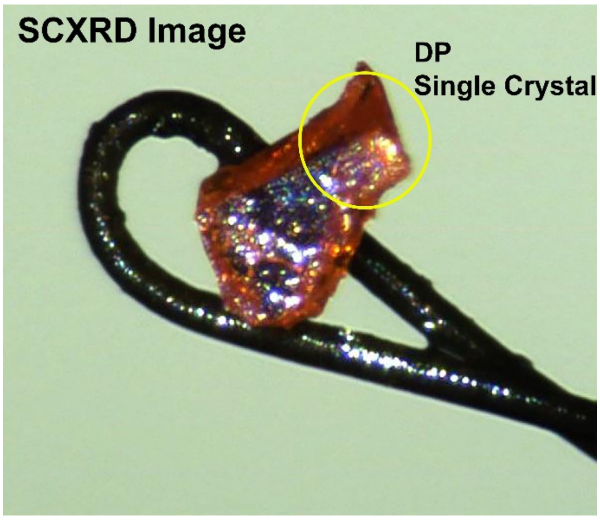
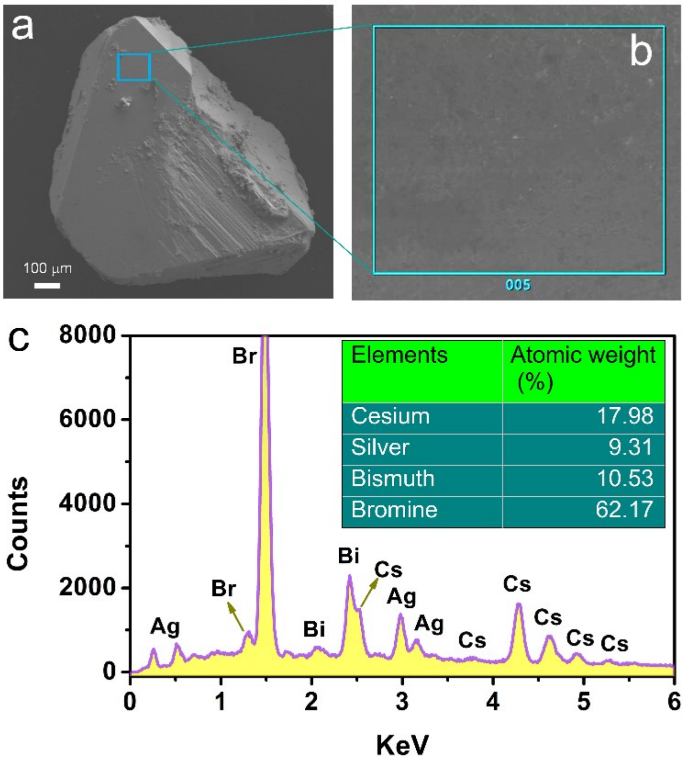


Fig. S1: Optical image of the DP single crystal mounted on the nylon loop with the help of silicon oil for SCXRD measurement.



Elements	Atomic weight (%)
Cesium	17.98
Silver	9.31
Bismuth	10.53
Bromine	62.17

Fig. S2: (a) FESEM image of the $Cs_2AgBiBr_6$ single crystal. **(b)** Magnified FESEM image of the marked area of $Cs_2AgBiBr_6$ single crystal. **(c)** Corresponding FESEM-EDX atomic spectra of $Cs_2AgBiBr_6$ single crystal. The inset shows the corresponding atomic percentage of each element.

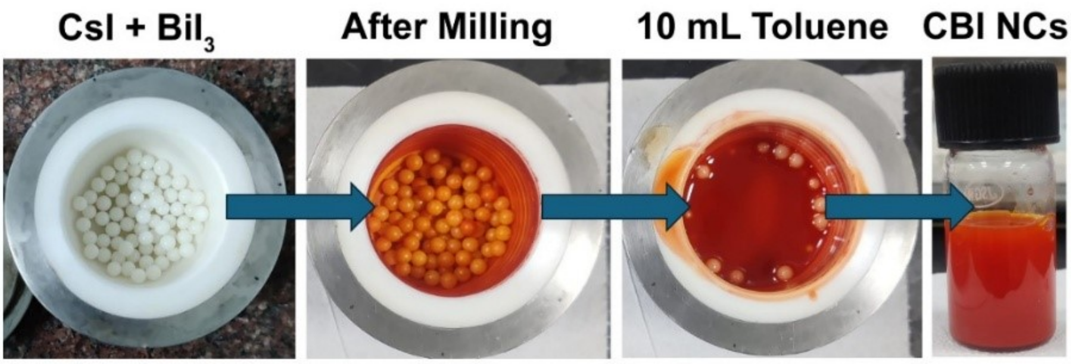
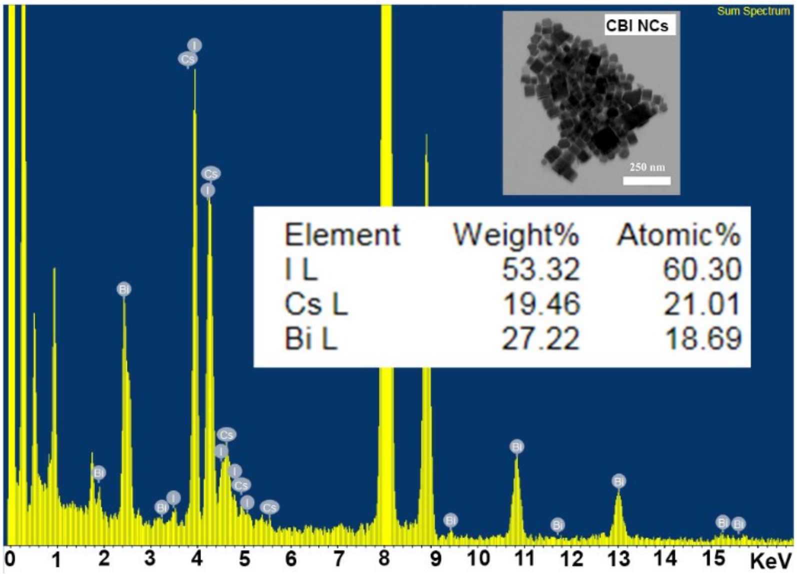


Fig. S3: Schematic illustration of the preparation method of $Cs_3Bi_2I_9$ NCs using the ball milling method.



Element	Weight%	Atomic%
I L	53.32	60.30
Cs L	19.46	21.01
Bi L	27.22	18.69

Fig. S4: FETEM-EDX atomic spectra of $Cs_3Bi_2I_9$ NCs.

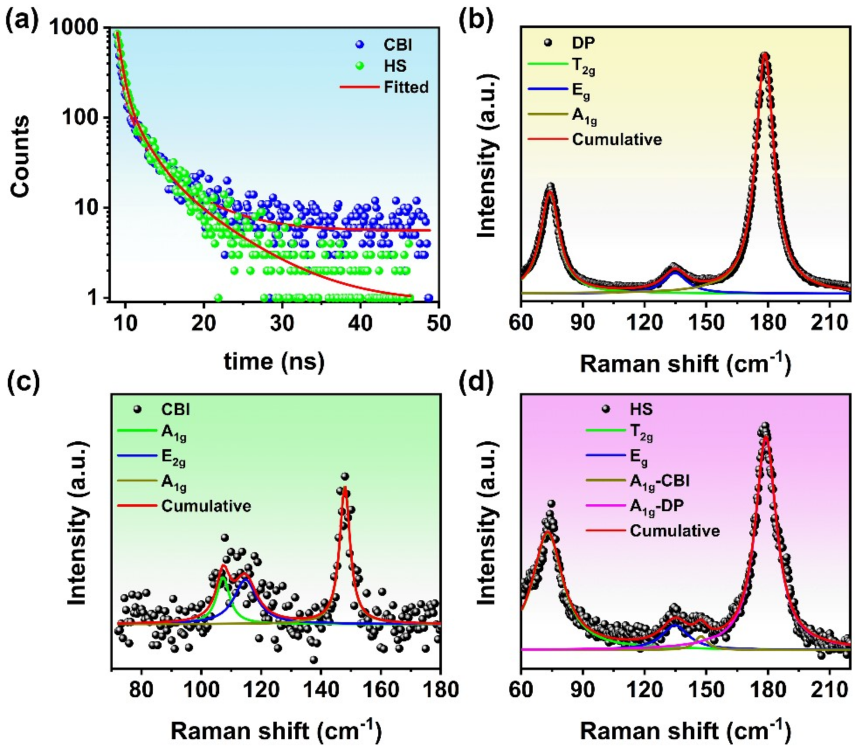


Fig. S5: (a) TRPL spectra of $Cs_3Bi_2I_9$ and $Cs_2AgBiBr_6/Cs_3Bi_2I_9$ HS. Deconvoluted Raman spectra of **(b)** $Cs_2AgBiBr_6$ SC, **(c)** $Cs_3Bi_2I_9$ NC thin film, and **(d)** $Cs_2AgBiBr_6/Cs_3Bi_2I_9$ heterostructure.

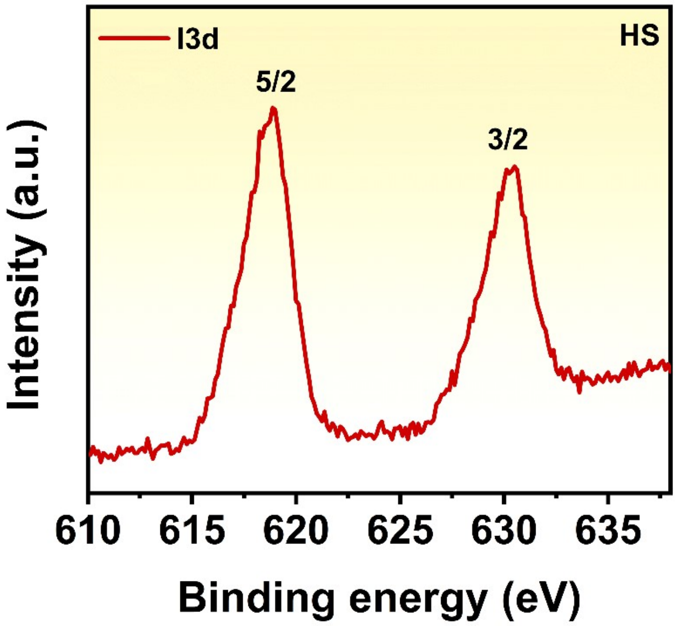


Fig. S6: XPS spectra of Iodine in the DPSC/CBI HS system.

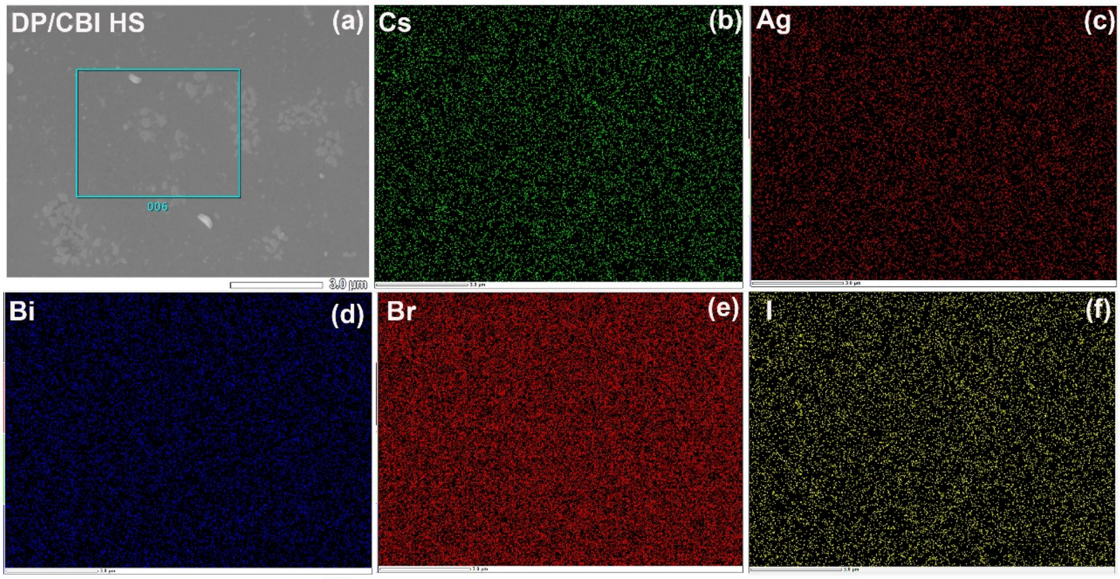


Fig. S7: (a) FESEM image of the DP/CBI HS. **(b-f)** FESEM-EDX mapping of all the constituent elements present in the HS.

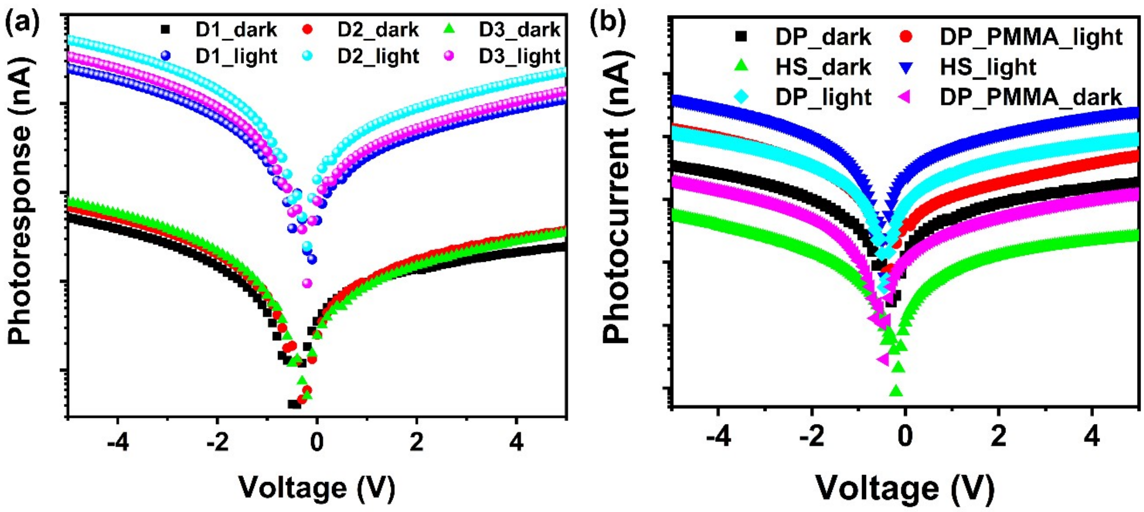


Fig. S8: (a) I-V characteristics of the multiple HS devices with the same configuration. **(b)** Comparison of the I-V characteristics of DP, DP-PMMA, and HS devices.

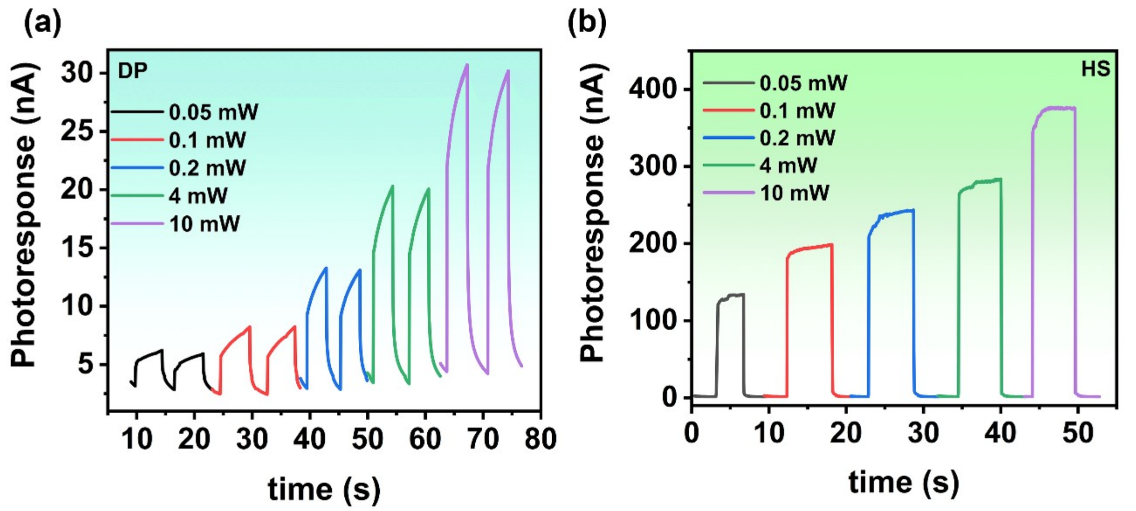


Fig. S9: Power-dependent photoresponse of the pristine and HS device under 450 nm laser illumination.

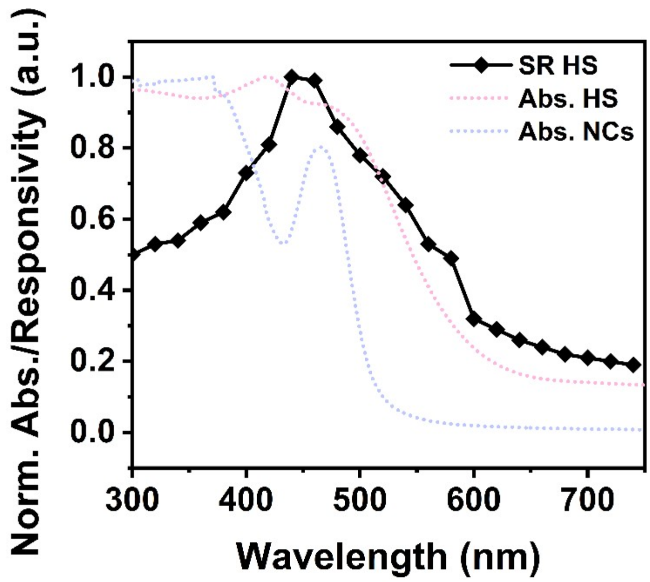


Fig. S10: Spectral responsivity of the HS device with reference to the absorbance of HS and CBI NCs.

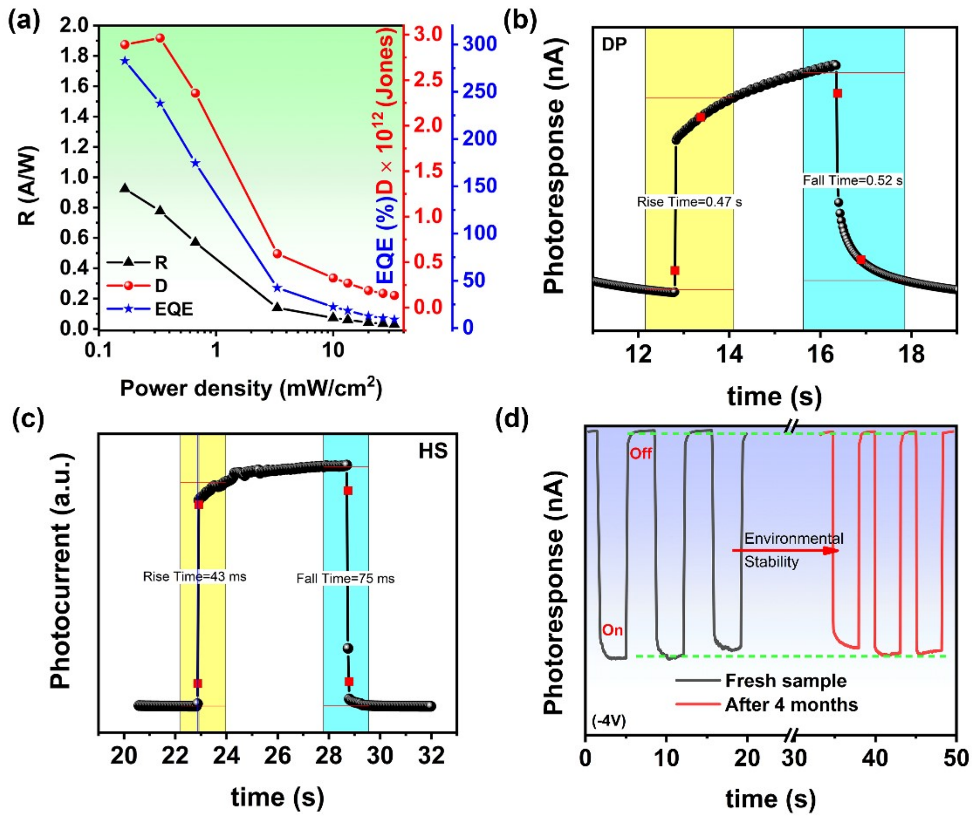


Fig. S11: (a) Comparison of responsivity, detectivity and external quantum efficiency of the $Cs_2AgBiBr_6/Cs_3Bi_2I_9$ heterostructure device. **(b, c)** rise and fall time of pristine $Cs_2AgBiBr_6$ SC and $Cs_2AgBiBr_6/Cs_3Bi_2I_9$ heterostructure device. **(d)** Stability performance of the HS device after 4 months of storage under ambient conditions.

Table S1: The unit cell parameters obtained from SCXRD measurement.

Parameters	Compound name
Formula	$Ag_2Bi_2Br_{12}Cs_4$
Mol.wt.	2124.26
Crystal System	Cubic
Space group	Fm-3m
a (Å)	11.264(6)
b (Å)	11.264(6)
c (Å)	11.264(6)
α (°)	90
β (°)	90
γ (°)	90
V ($Å^3$)	1429.3(2)
Density, gcm^{-3}	4.936
Abs. coeff., mm^{-1}	35.408
F (000)	1800
Total no. of reflections	110
Reflections, I > 2σ(I)	110
Max. θ/°	25.242
Ranges (h, k, l)	$-14 \leq h \leq 14$ $-14 \leq k \leq 14$ $-14 \leq l \leq 14$
Complete to 2θ (%)	98.9
Data / restraints / parameters	110/0/8
GooF (F^2)	1.265
R indices [I > 2σ(I)]	0.0112
wR_2 [I > 2σ(I)]	0.0258
R indices (all data)	0.0112
wR_2 (all data)	0.0258

Table S2: Peak positions of A_{1g} Raman mode in pristine and the HS materials obtained from the fitted Raman spectra in **Fig. S5**.

Material	A_{1g}-mode (peak position) (cm^{-1})
$Cs_2AgBiBr_6$ (DP)	178.93
$Cs_3Bi_2I_9$ (CBI)	147.44
$Cs_2AgBiBr_6/Cs_3Bi_2I_9$ HS	DP (A_{1g}) - 178.45 CBI (A_{1g}) - 147.93

Table S3: Stoichiometric ratio calculations obtained from deconvoluted XPS spectra of each element.

Element	Peak Area (A)	RSF (S)	Corrected Intensity (A/S)	Atomic %	Stoichiometric Ratio (Bi=1)
Cs	60835.42	7.041	8640.17	27.8	2.67
Ag	15819.38	5.987	2642.29	8.5	0.82
Bi	29549.46	9.140	3232.98	10.4	1.00
Br	14966.11	1.053	14212.83	45.7	4.40
I	14777.52	6.206	2381.17	7.7	0.74

Table S4: Comparison of the theoretical and measured composition of the DPSC/CBI HS from XPS data.

Composition	Cs	Ag	Bi	Br	I	Comment
Ideal $Cs_2AgBiBr_6$	2	1	1	6	0	No Iodide
Ideal $Cs_3Bi_2I_9$	1.50	0	1	0	4.50	No Silver, No Bromide
Theoretical (90:10) ($Cs_2AgBiBr_6$:$Cs_3Bi_2I_9$)	1.91	0.82	1	4.91	0.82	Expected for 90:10 mixture for Bi=1
XPS (Bi=1)	2.67	0.82	1	4.40	0.74	Measured surface composition