

Supporting information

Flexible and broadband colloidal quantum dots photodiode array for pixel-level X-ray to near-infrared image fusion

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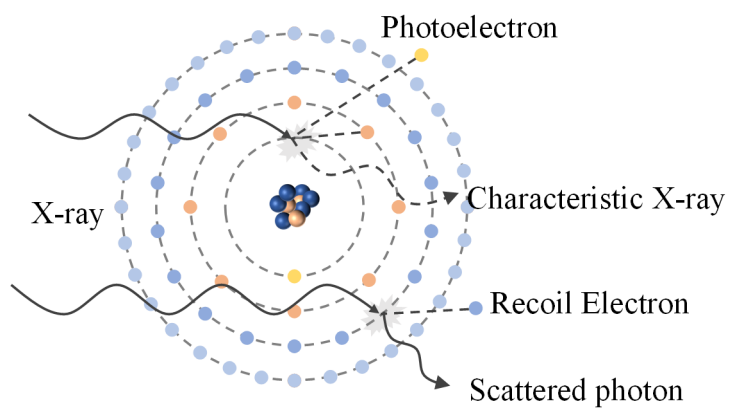


Fig. S1| Mechanism investigation of interactions between X-ray and PbS CQDs film.

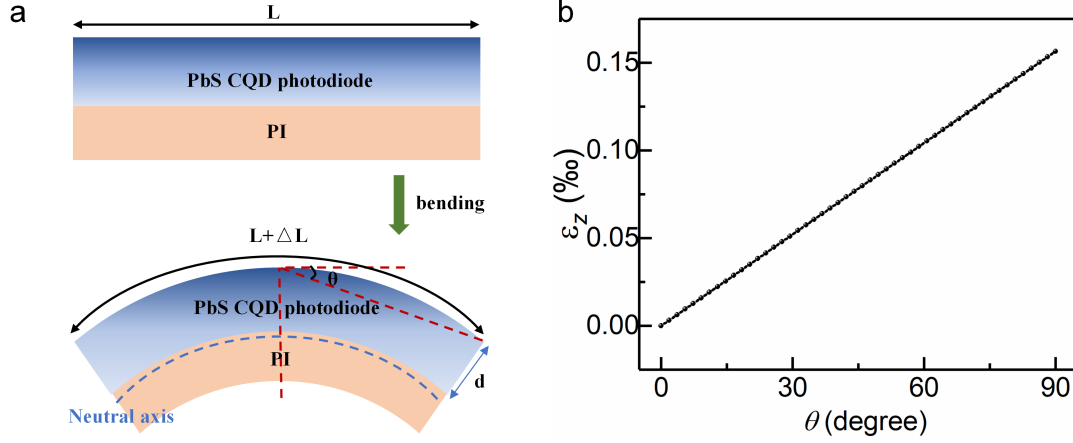


Fig. S2| a, Schematic diagram of bended PbS CQDs photodiode. **b**, Strain as a function of bending curvature.

Analysis of strain within a bended device is shown in Fig. S2a. As the PbS CQDs device is one thousand times thinner than the PI substrate¹, the neutral plane with zero strain is situated on the surface of the PI substrate¹. The strain ε_z at different positions can be rewritten as below

$$\varepsilon_z = \frac{z - z_{NA}}{r} \quad (S1)$$

where Z is the location of the CQDs device, Z_{NA} is the location of neutral plane, r is the curvature radius of film. r can be calculated by the equation:

$$r = \frac{360^\circ}{4\theta} \times \frac{l}{2\pi} \quad (S2)$$

where l is the length of the CQDs device, θ is the bending angle. Thus, we can obtain the relationship between θ and ε_z as shown in Fig. S2b. The maximum strain of PbS CQDs device is 0.15% at a bending angle of 90°. According to the previous report, the average inter-dot spacing is $\sim 3.21 \text{ nm}^2$. The inter-dot spacing of PbS CQDs only changes 0.00045 nm. It is possible to achieve a high degree of curvature even when the PbS CQDs are chemically bonded.

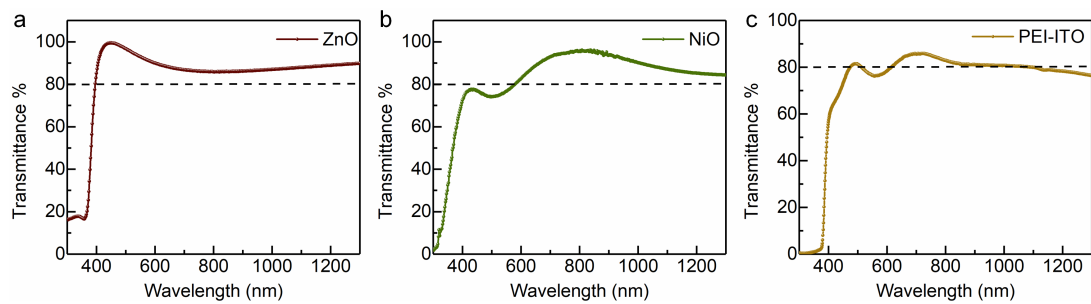


Fig. S3| Transmittance of flexible substrate and transport layers. a, Transmittance of ZnO film with thickness of 120 nm. **b,** Transmittance of NiO_x film with thickness of 40 nm. **c,** Transmittance of polystyrene naphthalate (PEN) with ITO layer.

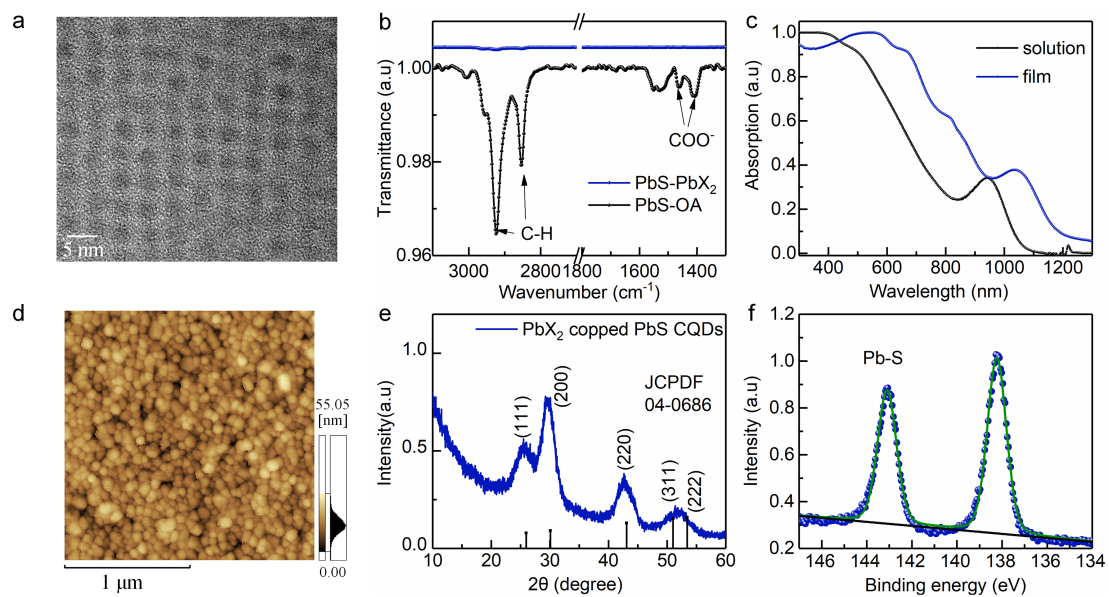


Fig. S4| Characterization of PbS CQDs. a, TEM image of PbS CQDs capped by oleic acid (CQD-OA). **b,** FTIR spectra of CQD-OA and PbX_2 ($X=\text{Br}, \text{I}$) capped PbS CQDs (CQD- PbX_2) film. **c,** Absorption spectra of CQD-OA solution and CQD- PbX_2 film. **(d)** AFM image, **(e)** XRD pattern and **(f)** XPS spectrum of CQD- PbX_2 film.

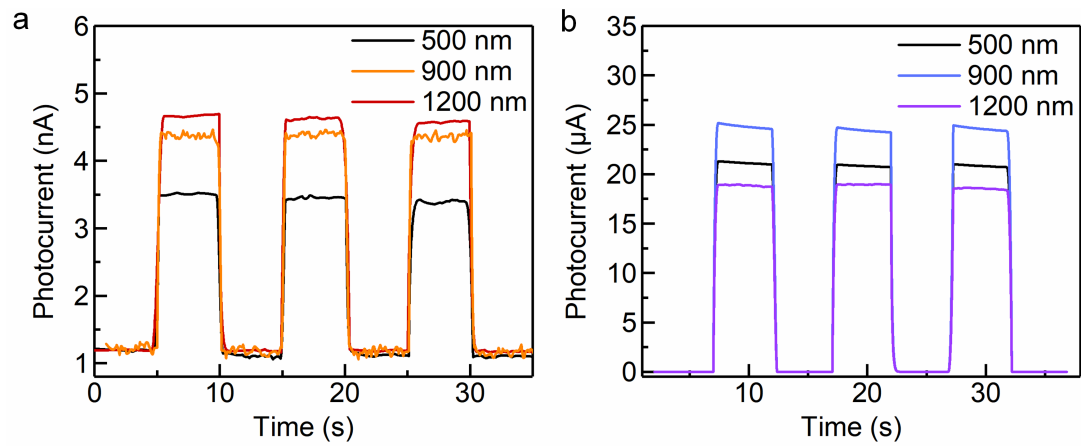


Fig. S5| Photoresponse of PbS CQDs photodiodes with different thickness of PbS CQDs layer. a, Transient responses at -0.1 V bias under $5.1 \text{ mGy}_{\text{air}} \text{ s}^{-1}$ dose rates X-ray. **b,** Transient responses at -0.1 V bias illuminated by 970 nm LED with a power density of 0.45 mW cm^{-2} .

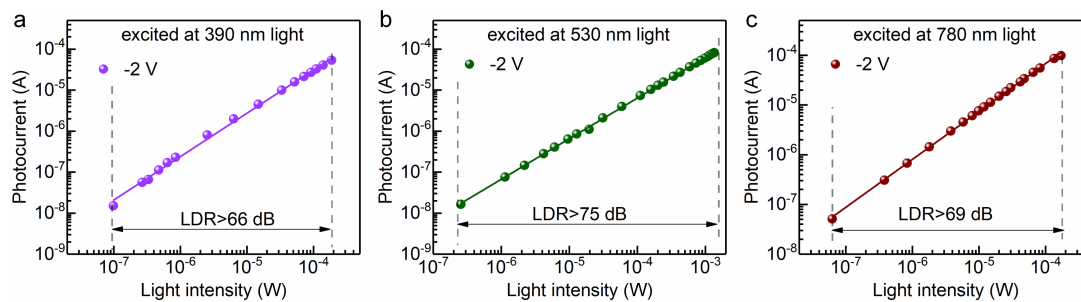


Fig. S6| Linear dynamic range (LDR) of PbS CQDs photodiode at the bias of -2 V under 390 nm (a), 530 nm (b) and 780 nm (c) illumination, respectively. As the photocurrent is linear within the test range, the LDR values are larger than 66 dB, 75 dB and 69 dB, respectively.

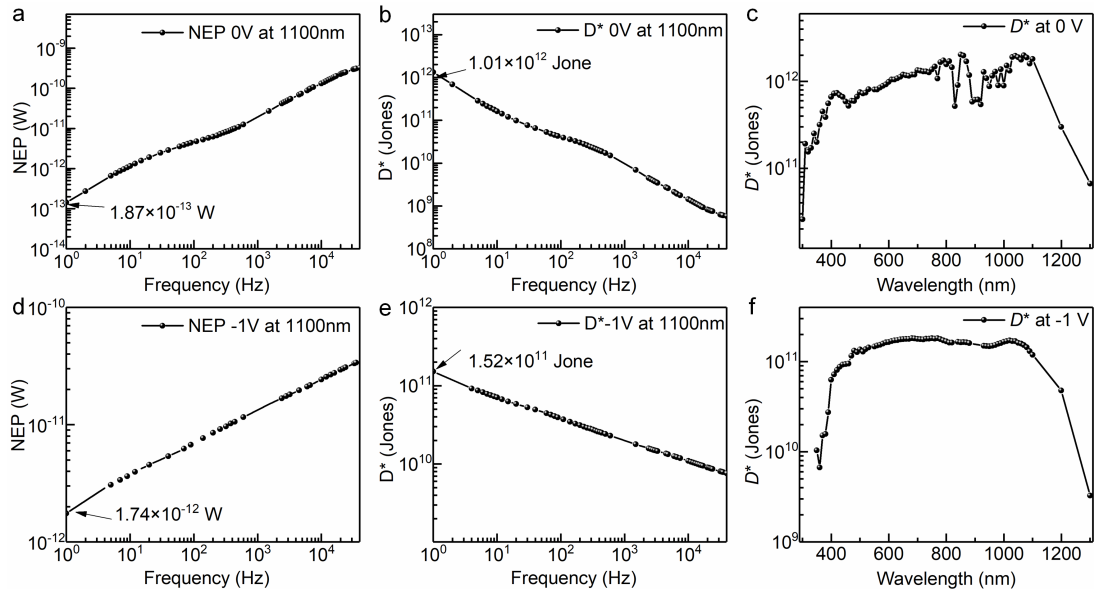


Fig. S7 | (a, b) Calculated NEP with normalized bandwidth. NEP is $1.87 \times 10^{-13} \text{ W Hz}^{-1/2}$ at zero bias and $1.74 \times 10^{-12} \text{ W}$ at -1 V , respectively. (d, e) Measured D^* as a function of frequency at zero bias and -1 V . D^* at 1 Hz is about $1.01 \times 10^{12} \text{ Jones}$ at zero bias and $1.52 \times 10^{11} \text{ Jones}$ at -1 V , respectively. (c, f) Measured D^* as a function of wavelength at zero bias and -1 V .

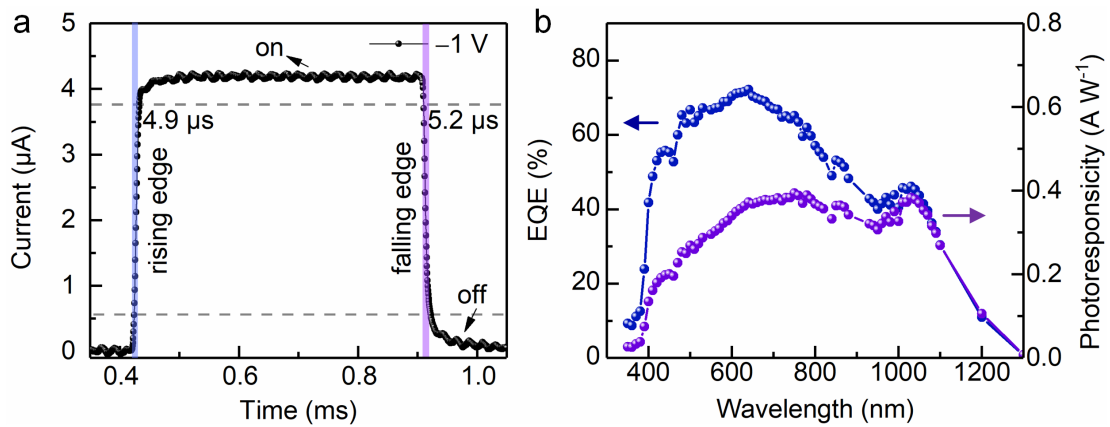


Fig. S8| Device performance of PbS CQDs photodiode under UV-Vis-NIR illumination. a, Temporal response of PbS CQDs photodiode at -1 V under 780 nm illumination. The rising and falling time are 4.9 and 5.2 μs . **b,** EQE and responsivity spectra of PbS CQDs photodiode at -1 V.

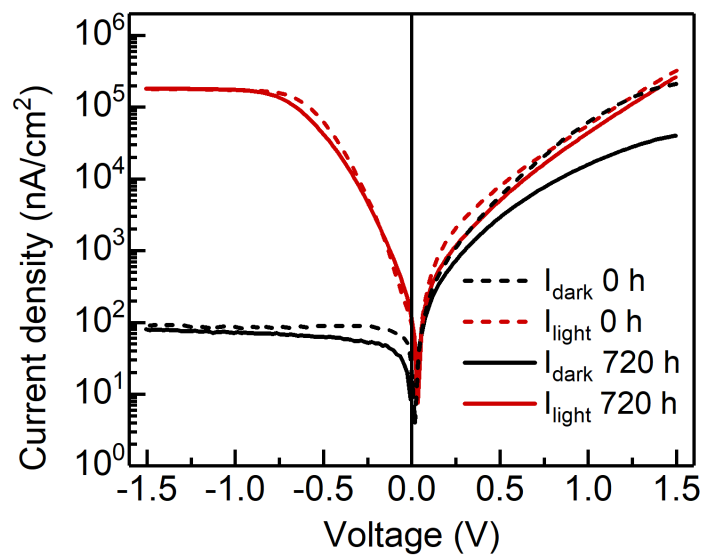


Fig. S9| Stability of PbS CQDs photodiode. The photocurrent and dark current density show negligible change after storing the device in dry air for 720 h.

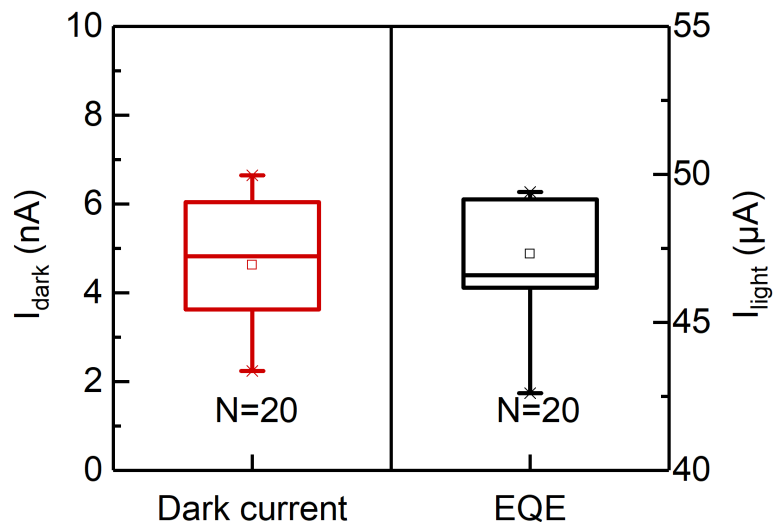


Fig. S10| Homogeneity of PbS CQDs photodiodes. N represents the number of device.

Table S1| Performance list of flexible photodetectors.

material	Spectral range (nm)	R (A/W ⁻¹)/ EQE (%)	Dark current density (nA/cm ²)	Detectivity (Jones)	Response time (s)	LD R (dB)	Ref.
Sb ₂ Se ₃	450–1050	0.42 83%	~900 (-0.1 V) ~2000 (-1 V) ^[C]	2.4×10 ¹¹ [M]	1.6×10 ⁻⁶	95	1
organic	350–1000	1200%	~50 (-1 V) ~100 (-5 V) ^[C]	2.0×10 ¹² [M]	/	158	3
MAPbI ₃	300–800	0.4 75%	~34 (-0.1 V)	1.1×10 ¹⁰ [M]	9.8×10 ⁻⁷	112	4
ZnO/PbS CQDs	350–1100	4.54	/	3.98×10 ¹² [C]	1.01	>60	5
PbS CQDs	350–1300	0.38 76.6%	12.6 (-0.1 V) 50.9 (-1 V)	1.01×10¹² [M]	5×10⁻⁶	>85	This work

‘C’ means the dark current density calculated from the given dark current and device area in the article.

Table S2| The EDS spectra detected from an area of the PbS CQDs film.

Element	Line type	Mass percent (%)	Atomic percent (%)
Pb	M	64.78	38.46
I	L	20.51	19.88
S	K	8.28	31.77
Br	L	6.42	9.89

Table S3| The key parameters of flexible X-ray detectors.

Materials	Volume Sensitivity ($\mu\text{C mGy}^{-1} \text{cm}^{-3}$)	Bias (V)	Energy (KeV)	Year
PTAA-Bi ₂ O ₃	0.2	200	17.5	2012 ⁶
P8T2	0.158	50	17	2009 ⁷
TIPS-pentacene	72	0.2	17	2016 ⁸
Cs ₂ AgBiBr ₆	4	400	45	2018 ⁹
Bi ₂ O ₃	1712	10	50	2018 ¹⁰
Ga ₂ O ₃	271	50	40	2019 ¹¹
MAPb(I _{0.9} Cl _{0.1})I ₃	362	12	60	2020 ¹²
Cs _{0.1} (FA _{0.83} MA _{0.17}) _{0.9} Pb(Br _{0.17} I _{0.83}) ₃	55.8	0.1	70	2020 ¹³
SCU-13	13	100	80	2020 ¹⁴
Cs ₄ PbI ₆	305	10	30	2021 ¹⁵
FAPbI ₃	284.2	0.5	35	2021 ¹⁶
Cs ₂ TeI ₆	217	5	20	2021 ¹⁷
Ni-DABDT	19.72	1	26	2021 ¹⁸
DABCO-CsBr ₃	0.533	200	40	2022 ¹⁹
Cs ₂ TeI ₆	1512	10	29	2022 ²⁰
MA ₃ Bi ₂ I ₆	206.5	200	30	2022 ²¹
MPAZE-NH ₄ I ₃ ·H ₂ O	49.38	20	22	2023 ²²
PbS CQDs	200	0.1	50	This work

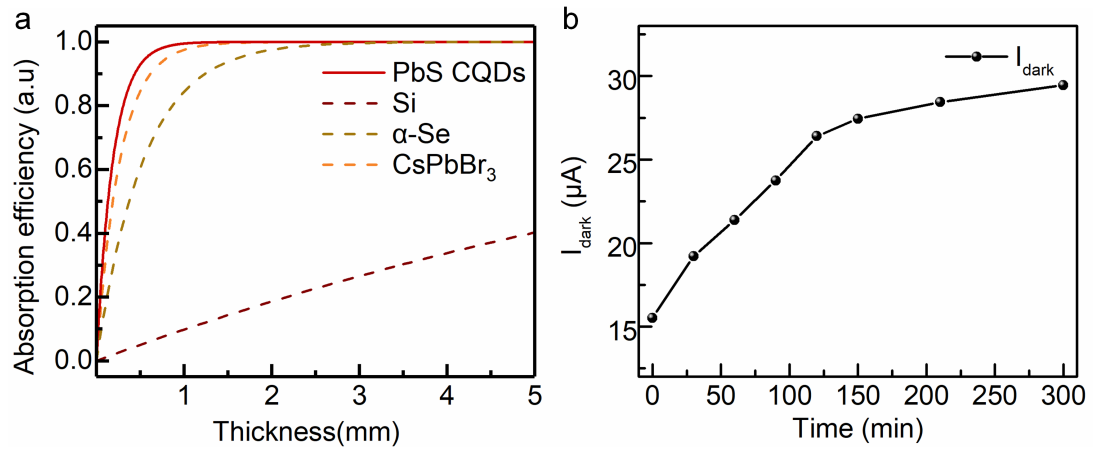


Fig. S11 | **a**, Absorption efficiency of a few semiconductors to 50 keV X-ray photon versus thickness. **b**, Irradiation stability of PbS polycrystalline film prepared by CBD (chemical bath deposition).

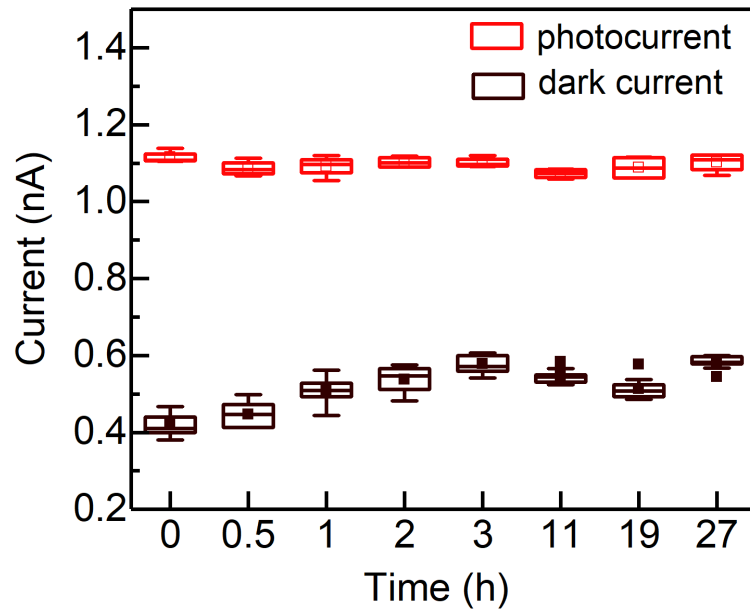


Fig. S12| Stability of PbS CQDs photodiodes under X-ray irradiation.

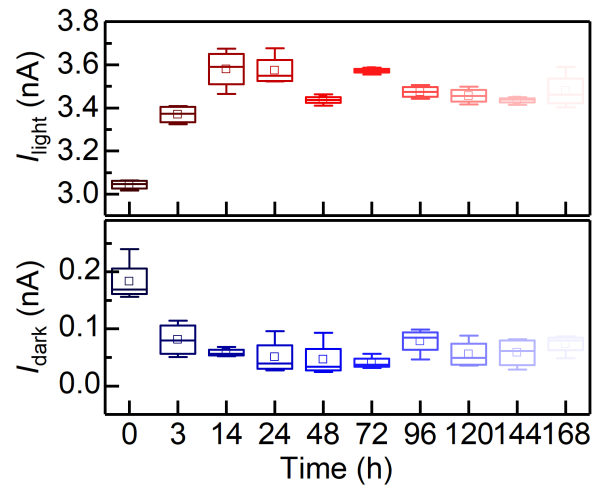


Fig. S13| Stability of PbS CQDs film under X-ray irradiation.

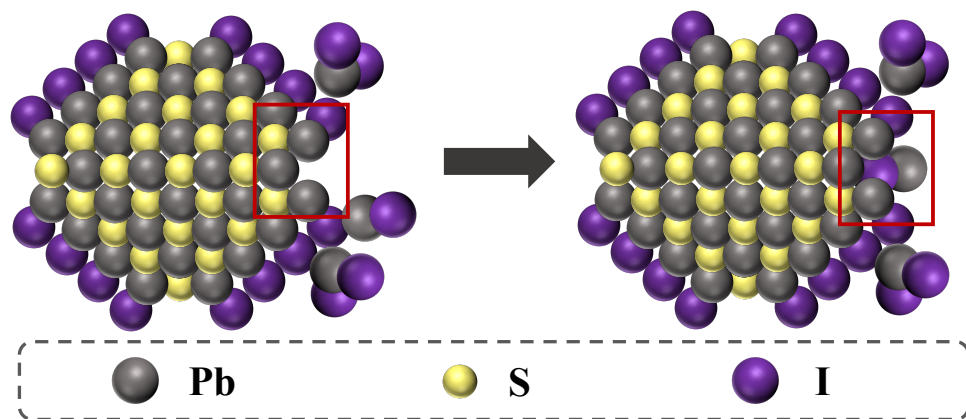


Fig. S14| Schematic diagram of the ligand migration under X-ray irradiation.

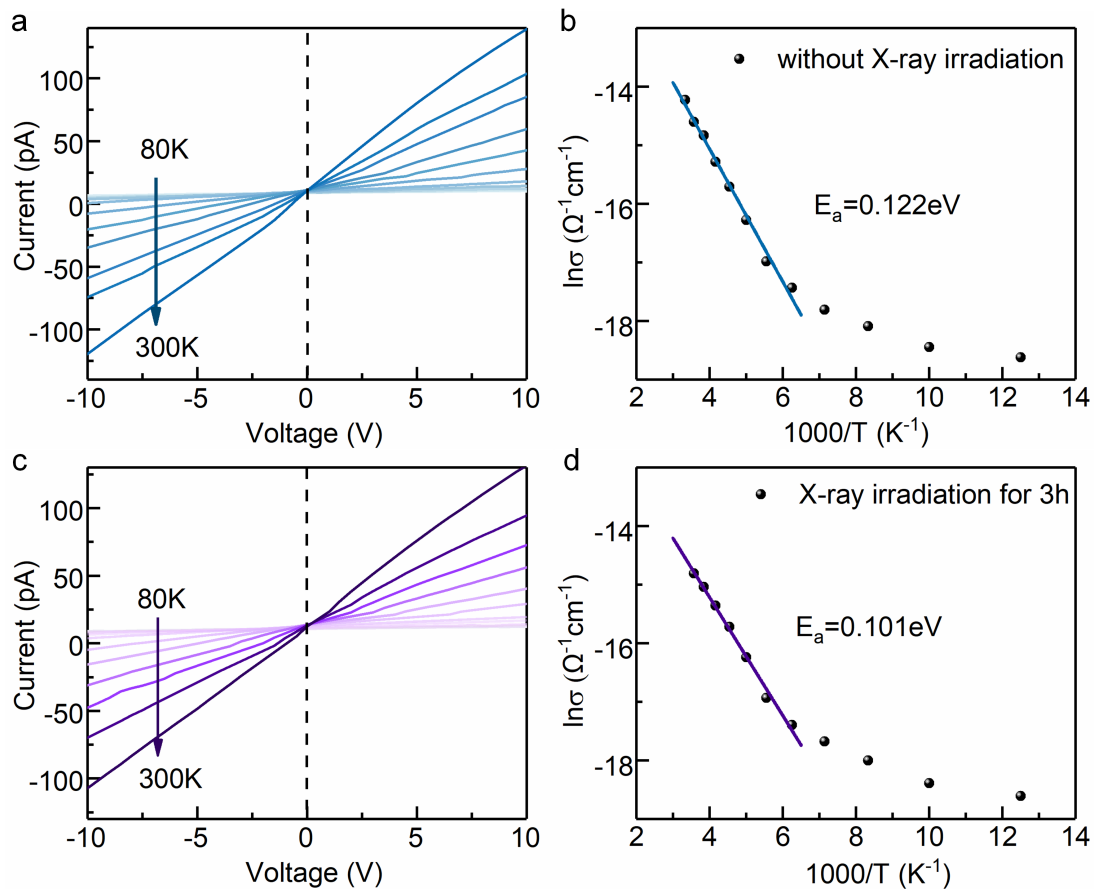


Fig. S15| Defect analysis of PbS CQDs film. (a, c) Current-voltage ($I-V$) curves of Au/PbS CQDs/Au photoconductor without and with X-ray irradiation from 80 K to 300 K, respectively. (b, d) Defect depth of PbS CQDs film without and with X-ray irradiation.

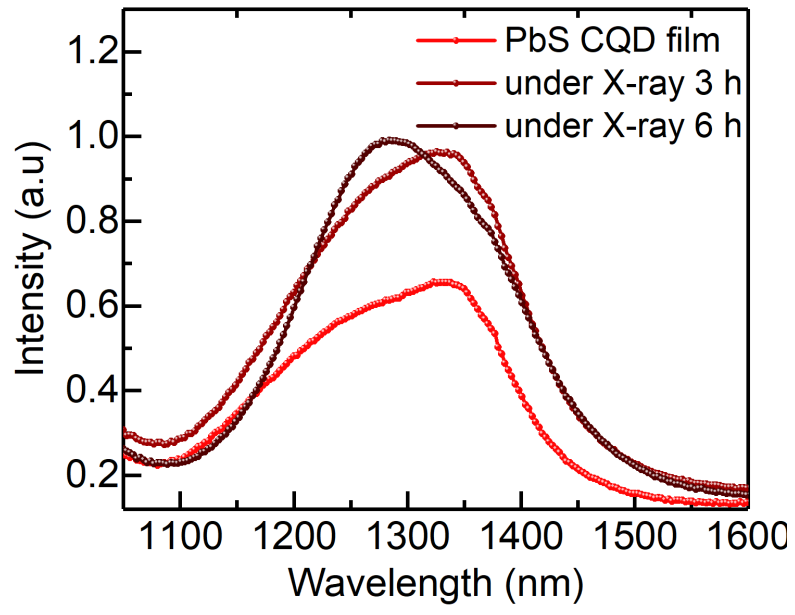


Fig. S16| Photoluminescence of PbS CQDs film under X-ray irradiation.

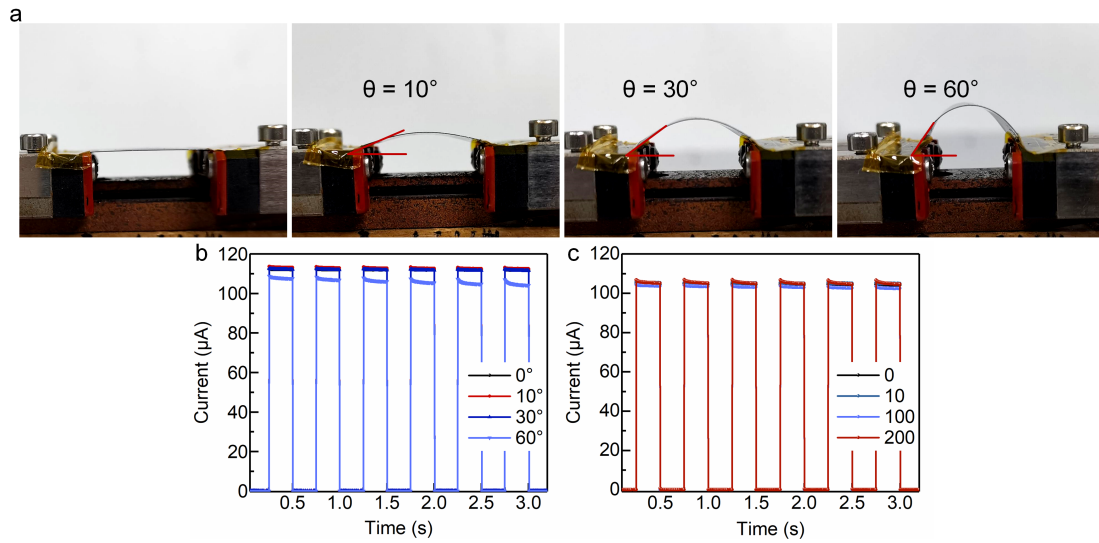


Fig. S17| Flexibility performance of PbS CQDs photodiode. a, Photographs of bended PbS CQDs photodiode array at varied angles from 0° to 60° . Photoresponse of flexible PbS CQDs photodiode at different bending angles (**b**) and different bending cycles at a fixed angle of 30° (**c**).

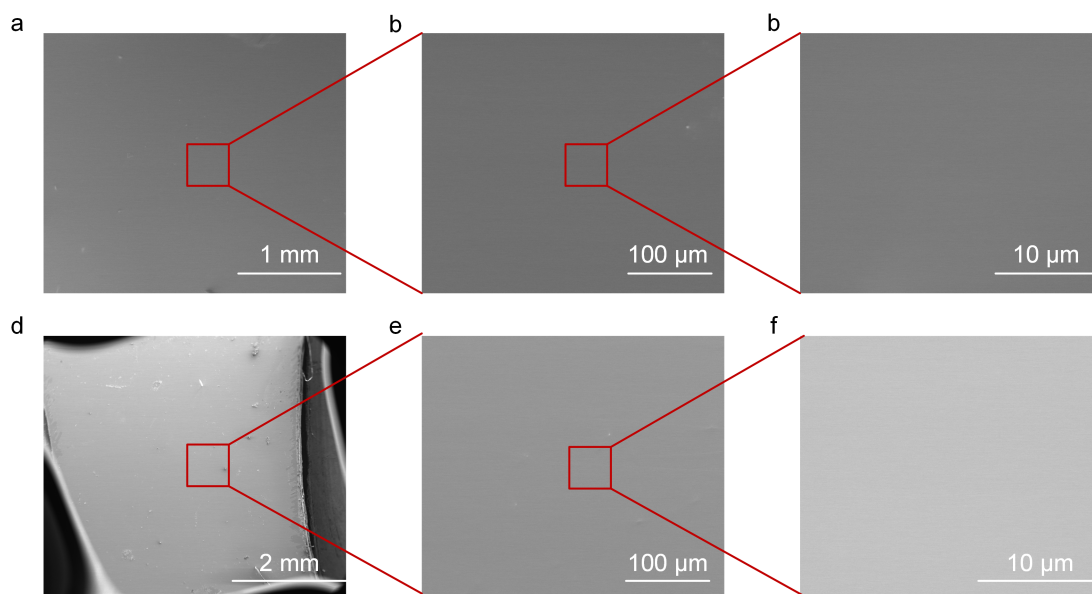


Fig. S18| Flexibility of PbS CQDs film. (a, b, c) SEM images of PbS CQDs films before bending with scar bars of 1 mm, 100 μm and 10 μm. (d, e, f) SEM images of PbS CQDs films under 30° bending with scar bars of 2 mm, 100 μm and 10 μm.

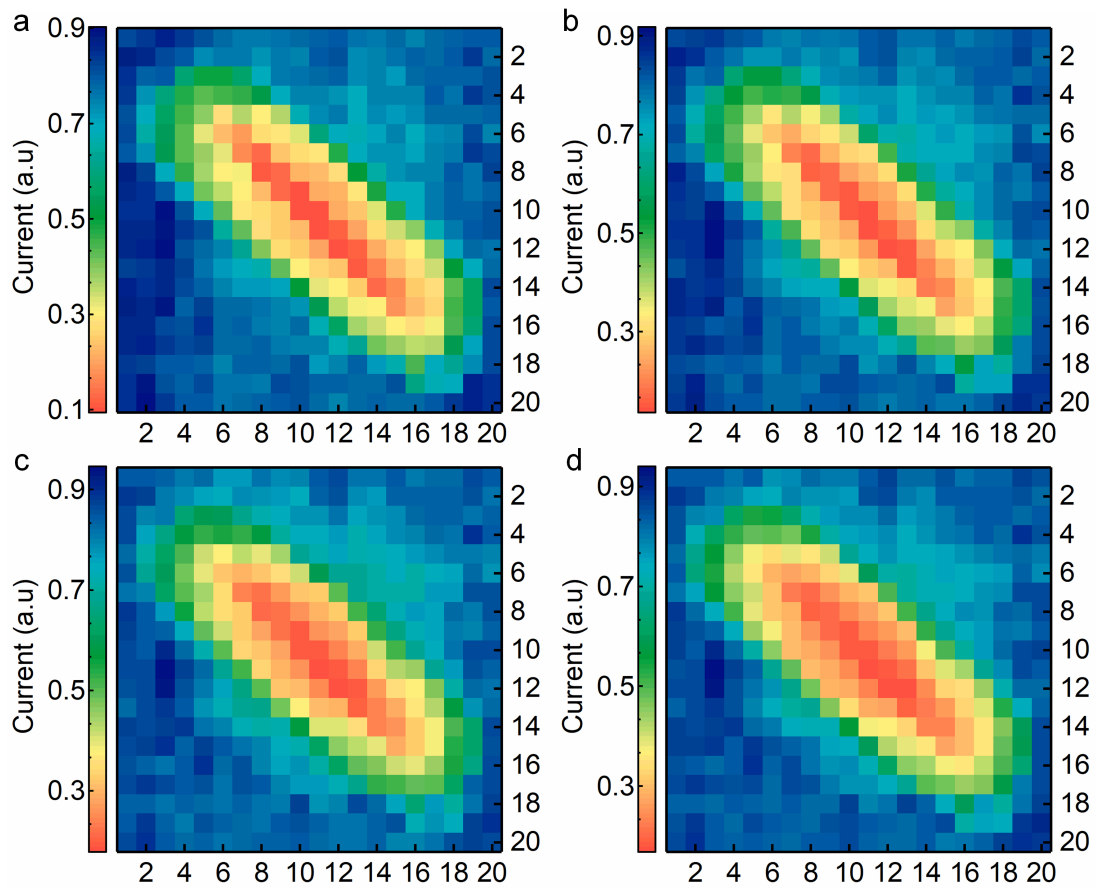


Fig. S19| Fused images with different weights coefficients. a, The weight values of X-ray image, visible image and NIR image are all 0.3. **b,** The weight values of X-ray image, visible image and NIR image are 0.25, 0.25 and 0.5 respectively. **c,** The weight values of X-ray image, visible image and NIR image are 0.16, 0.16 and 0.68 respectively. **d,** The weight values of X-ray image, visible image and NIR image are 0.25, 0.125 and 0.625 respectively.

Supplementary References

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