Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Exploring centimeter-sized single crystal of 2D Dion-Jacobson halide perovskite toward efficient X-ray detection

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Experimental Section

Synthesis. **1** was yielded from hydroiodic acid solution (55%~58%, Aladdin) containing stoichiometric amounts of $Pb(CH_3COO)_2 \cdot 3H_2O$ (99.5%, Aladdin) and (S)-1-(4-Nitrophenyl)ethanamine hydrochloride (98% Aladdin). The saturated solution was heated and stirred at 373 K for 20 min to obtain the clarified solution. Bulk single crystals (SCs) of **1** were grown from saturated solution at 0.5 K/day by temperature cooling method.

Powder X-ray diffraction (PXRD). PXRD of **1** was recorded in an atmospheric environment with a Rigaku MiniFlex diffractometer. Using a step size of 0.02° in the 2θ range of 5°-40°, the diffraction patterns were collected.

Thermogravimetric analysis (TGA) measurement. TGA was performed on STA449C Thermal Analyzer at temperatures ranging from room temperature to 800°C.

Single-crystal structure determination. On a Bruker 8 diffractometer, single-crystal X-ray diffractions of **1** was carried out using Mo Kα radiation at 150 K. The Crystalclear software program was used to analyze the data. Direct techniques were used to solve the structures of **1**, and the SHELXLTL software was used to refine them using full-matrix least-squares refinements on F^2.

UV–Vis–NIR Diffuse Reflection Spectroscopy. On a PE Lambda 900 UV-Visible spectrometer, the solid UV absorptions were measured at room temperature.

Computational description. Single-crystal structure data of **1** at 150 K was used for the theoretical calculations. Band structure and partial density of states (PDOS) were performed by the DFT method within the total-energy code CASTEP.

X-ray detection. The *I-V* traces and *I-t* curves under X-ray irradiation was also recorded using the 6517B high precision electrometer (Keithley, USA). A commercially available Ag target X-ray tube with X-ray photons energy up to 50 keV and peak intensity at 22 keV was used as the X-ray source (4 W, Mini-X2, Amptek, USA). The dosage rate of X-ray tube was modulated by altering its tube current and measured by a commercial X-ray dosimeter (Accu-Gold, Radcal, USA) attached with the ion chamber (10×6-180 model) in an integrating mode.

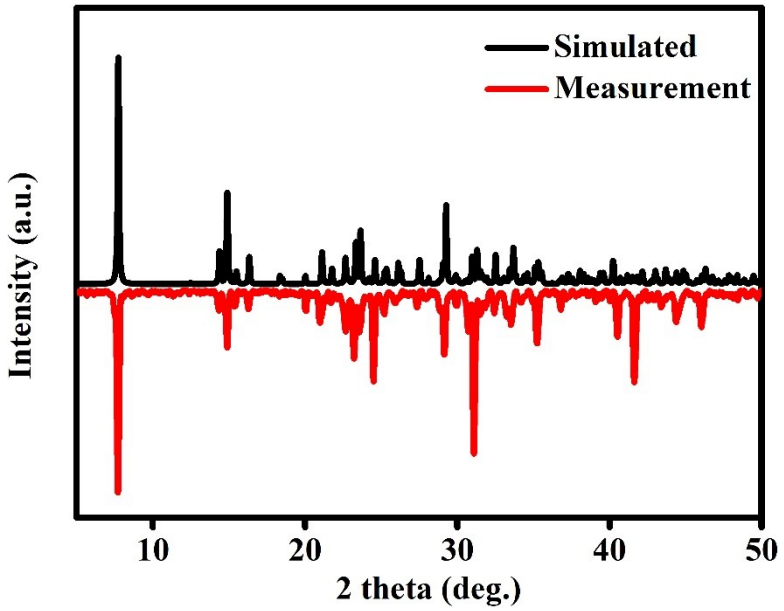

Fig. S1 The simulated and experimental PXRD patterns of **1**.

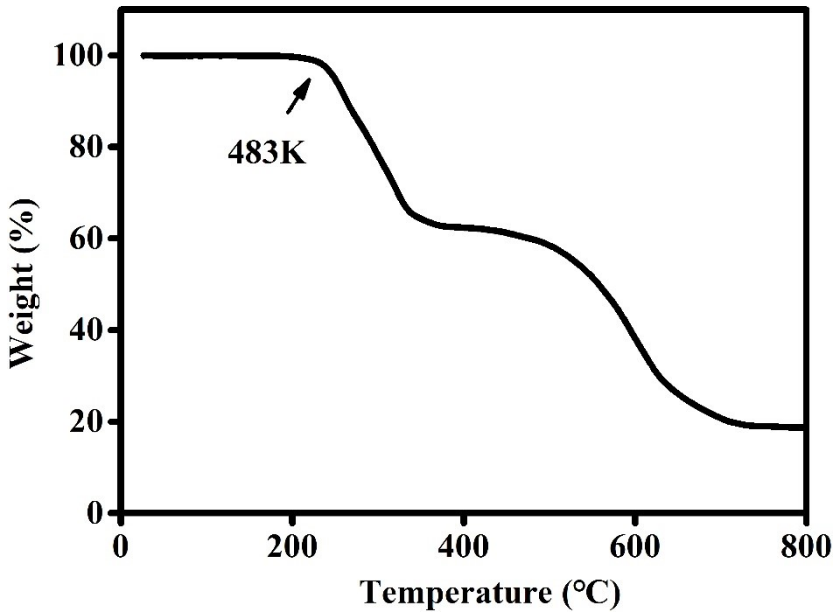

Fig. S2 Thermogravimetric analysis of synthesized **1**.

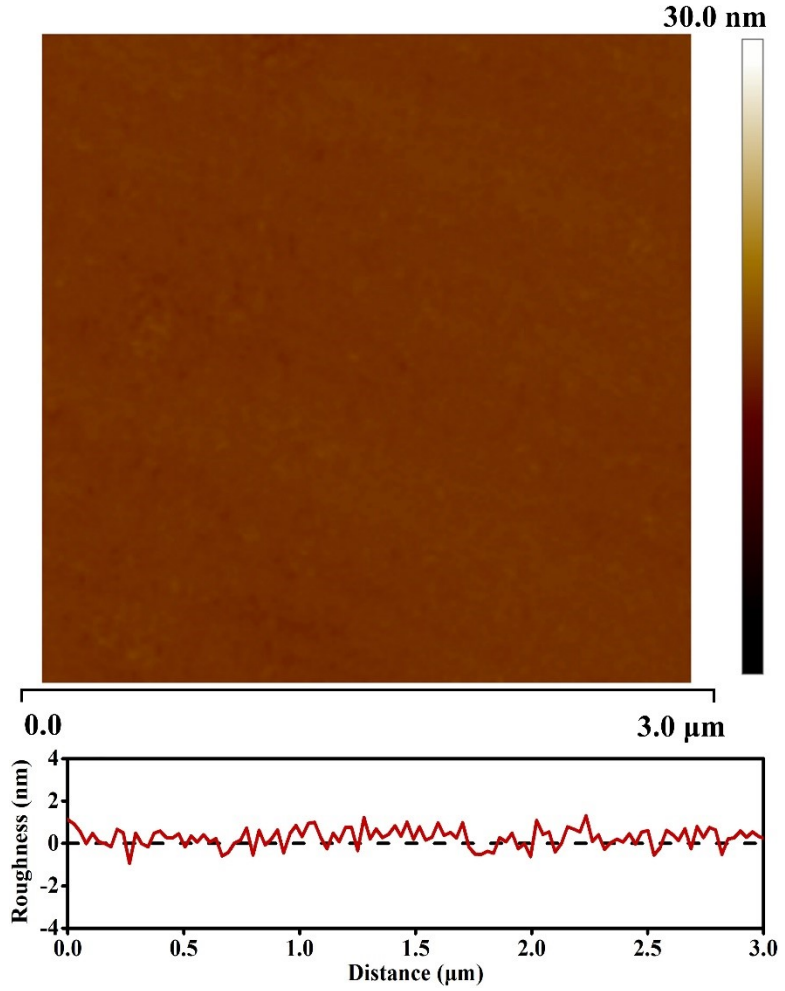

Fig. S3 Surface morphology measured by AFM.

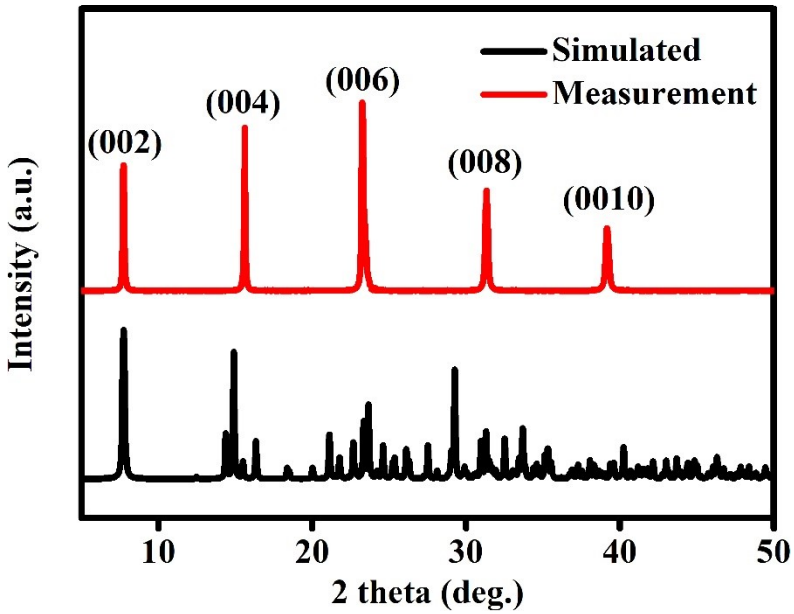

Fig. S4 XRD scan of the top facet of **1** SC.

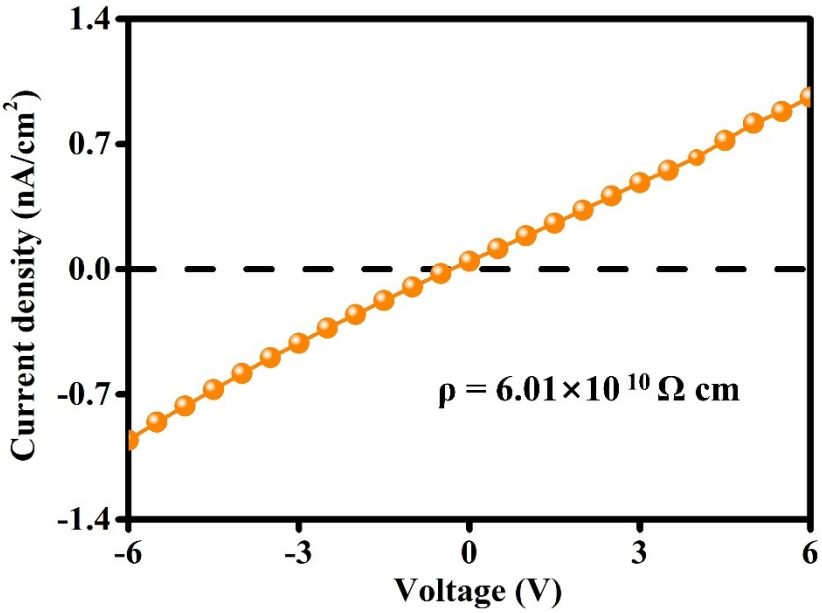

Fig. S5 Bulk resistivity of **1** SC.

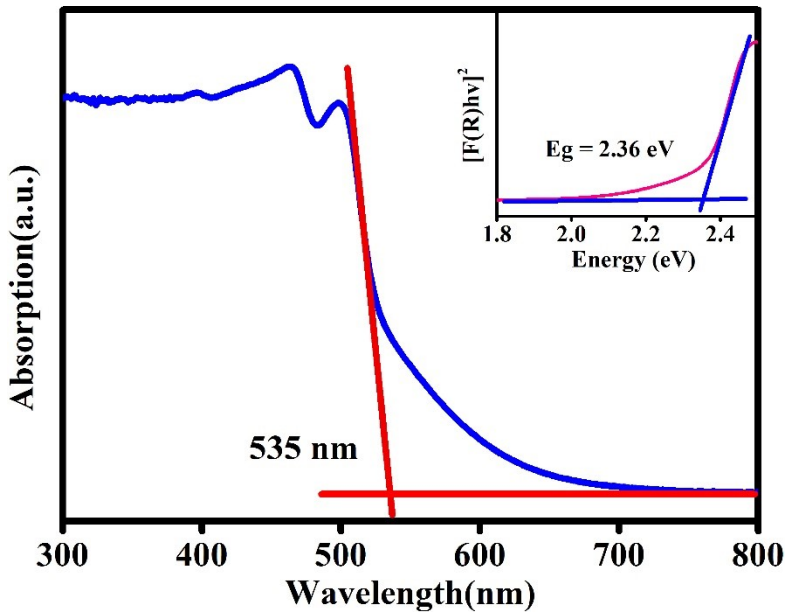

Fig. S6 The absorption spectrum and derived optical bandgap (inset) of **1**.

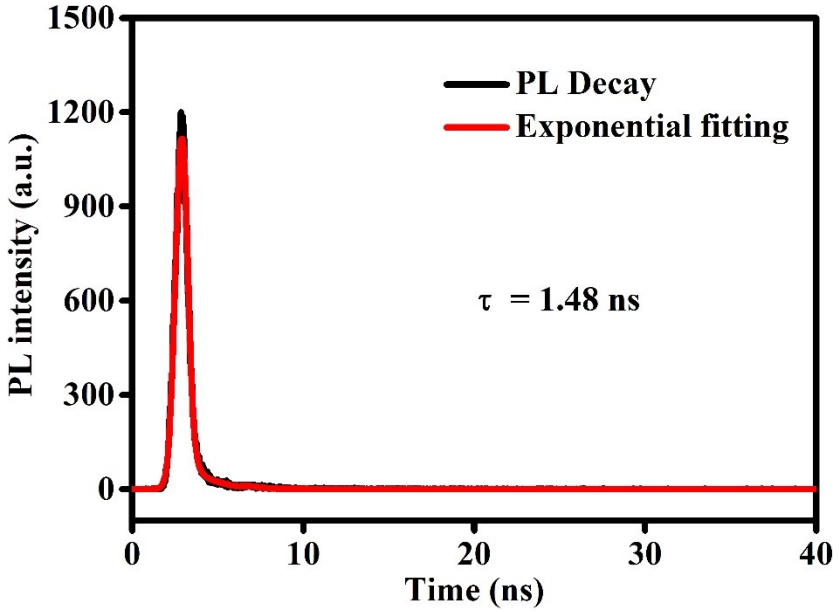

Fig. S7 PL decay curve monitored at 505 nm of **1** SC under 375 nm excitation.

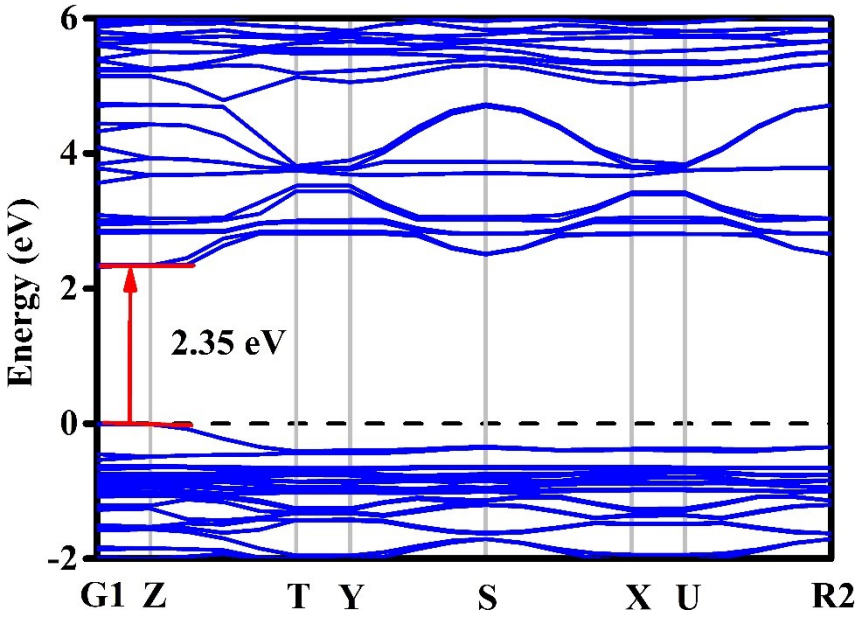

Fig. S8 Calculated band structure of **1.**

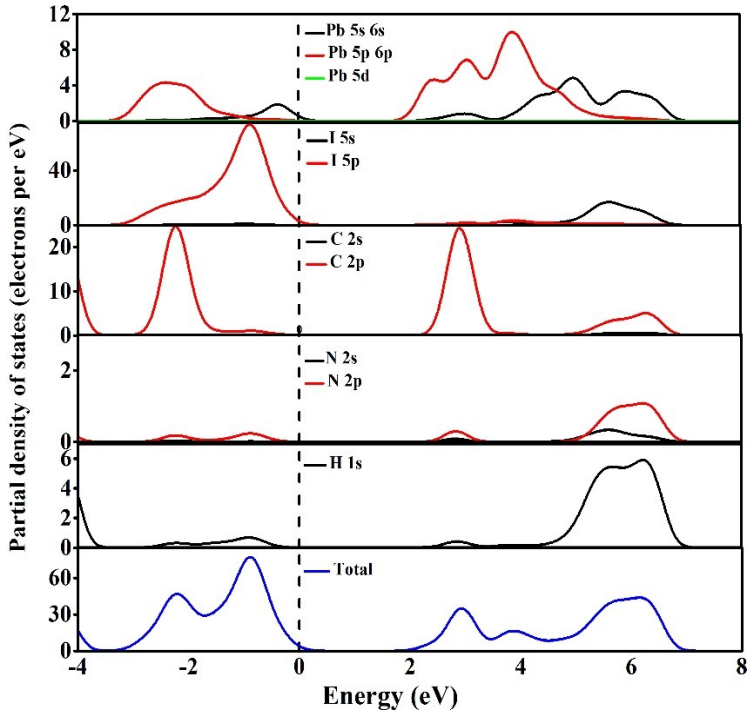

Fig. S9 PDOS spectra of **1**.

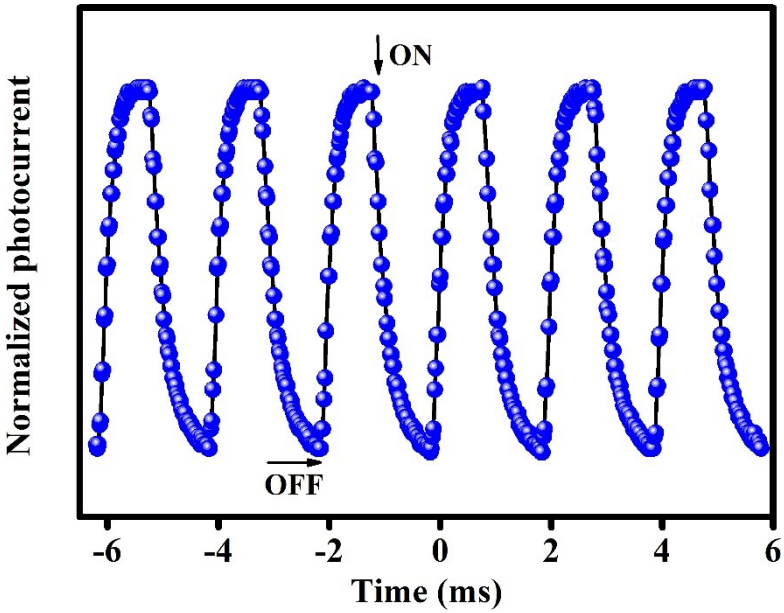

Fig. S10 Repetitive switching cycles of photoresponse for **1**.

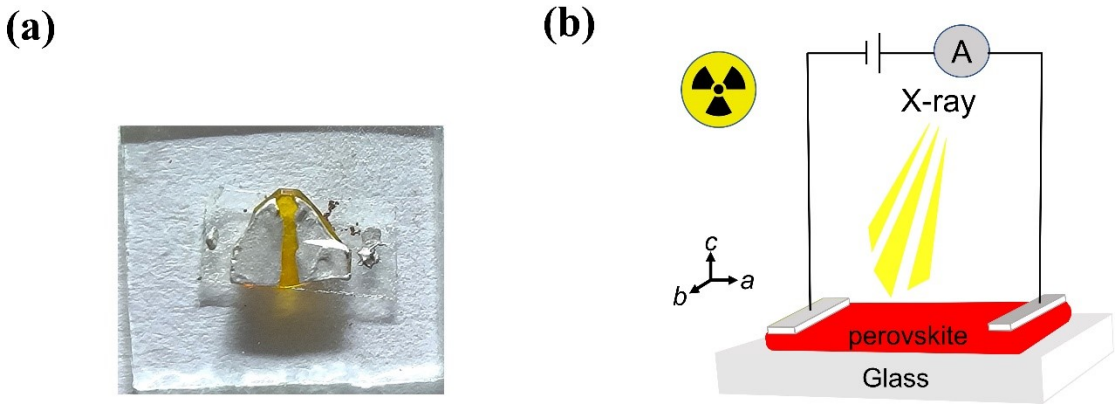

Fig. S11 (a) Device picture based on **1** SC. (b) Schematic illustration of X-ray detection based on **1** SC.

Table S1. Crystallographic data and structure refinements of **1** at 150 K.

Empirical formula	$C_8H_{14}I_4N_2Pb$
Formula weight	853.00
Temperature/K	150
Crystal system	orthorhombic
Space group	$P2_12_12_1$
a/Å	8.4311(3)
b/Å	9.0270(4)
c/Å	22.8714(12)
α/°	90
β/°	90
γ/°	90
Volume/ Å^3	1740.69(13)
Z	4
ρ_{calc}g/cm^3	3.255
μ/mm^{-1}	16.766
F(000)	1480.0
Radiation	MoKα(λ=0.71073)
2θ range for data collection/°	4.852 to 54.98
Reflections collected	10425
Data/restraints/parameters	3900/0/139
Goodness-of-fit on F^2	0.969
Final R indexes [$I \geq 2\sigma$ (I)]	R_1 = 0.0321, wR_2 = 0.0694
Final R indexes [all data]	R_1 = 0.0348, wR_2 = 0.0710
Largest diff. peak/hole/e Å^{-3}	1.21/-1.76

Table S2. A detection limit summary of perovskite single-crystal X-ray detectors.

Materials	Operating voltage (or electric field)	Detection limit	detection sensitivity	Ref.
$FA_3Bi_2I_9$	180 V	0.2 $\mu Gy_{air}\ s^{-1}$	598 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	1
$(BDA)PbI_4$	10 V	430 $nGy_{air}\ s^{-1}$	242 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	2
(R)-$(H_2MPz)BiI_5$	50 V mm^{-1}	4.38 $\mu Gy_{air}\ s^{-1}$	242 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	3
$MAPbBr_3$	-20 V	0.48 $\mu Gy_{air}\ s^{-1}$	2975 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	4
$(3AP)PbCl_4$	5 V	1.54 $\mu Gy_{air}\ s^{-1}$	791 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	5
$Cs_2AgBiCl_6$	2 V mm^{-1}	241 $nGy_{air}\ s^{-1}$	325 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	6
$DABCO\text{-}N_2H_5Br_3$	10 V	2.68 $\mu Gy_{air}\ s^{-1}$	1143 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	7
$(BA)_2PbI_4$	10 V mm^{-1}	241 $nGy_{air}\ s^{-1}$	148 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	8
α-Se	200 V	5.5 $\mu Gy_{air}\ s^{-1}$	20 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	9
1	5 V bias	236 $nGy_{air}\ s^{-1}$	270 $\mu C\ Gy_{air}^{-1}\ cm^{-2}$	**This work**

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