

## Supporting Information

### Centimeter-Sized Single Crystals of 2D Hybrid Perovskite toward Ultraviolet

#### Photodetection with Anisotropic Photoresponse

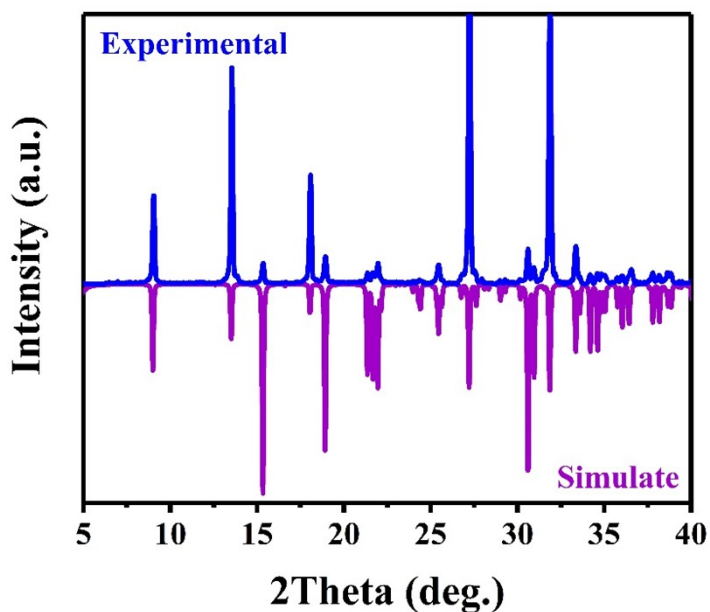
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### Figures



**Figure S1.** Experimental and calculated powder X-ray diffraction patterns of (BA)<sub>2</sub>CsPb<sub>2</sub>Br<sub>7</sub> at room temperature.

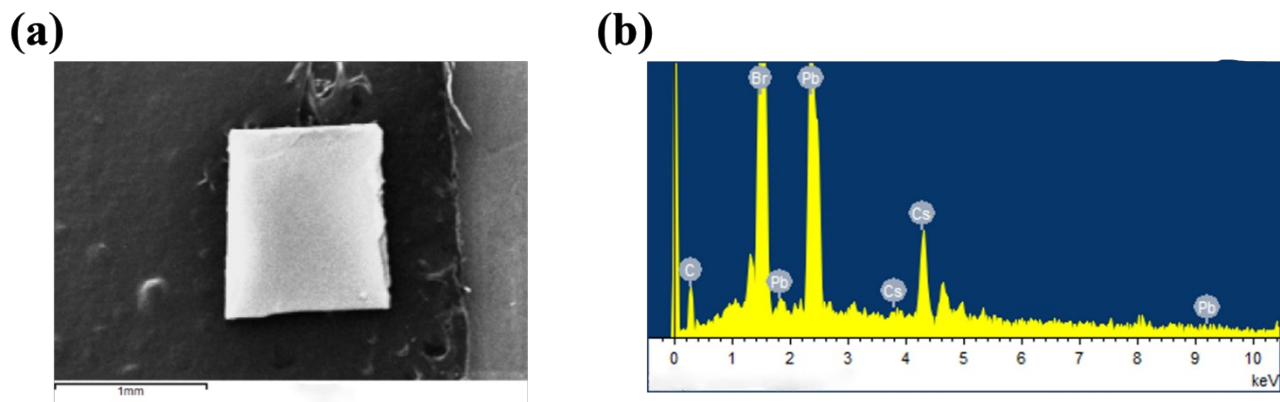


Figure S2. (a) Image and (b) EDS patterns of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$  single crystal.

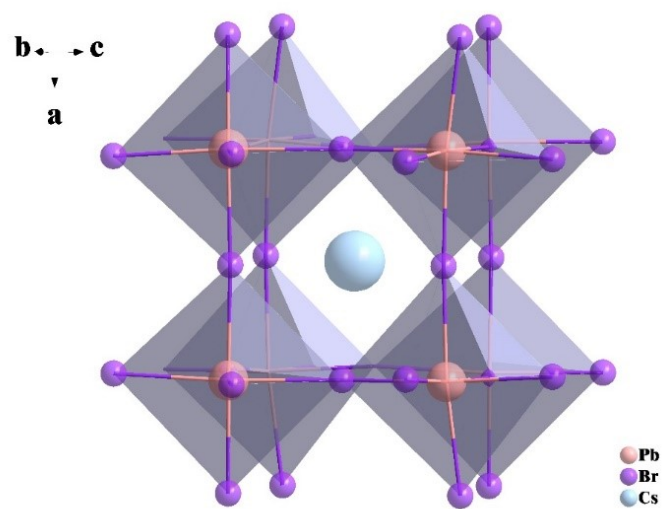


Figure S3. Crystal structures of cubic 3D perovskite  $\text{CsPbBr}_3$ .

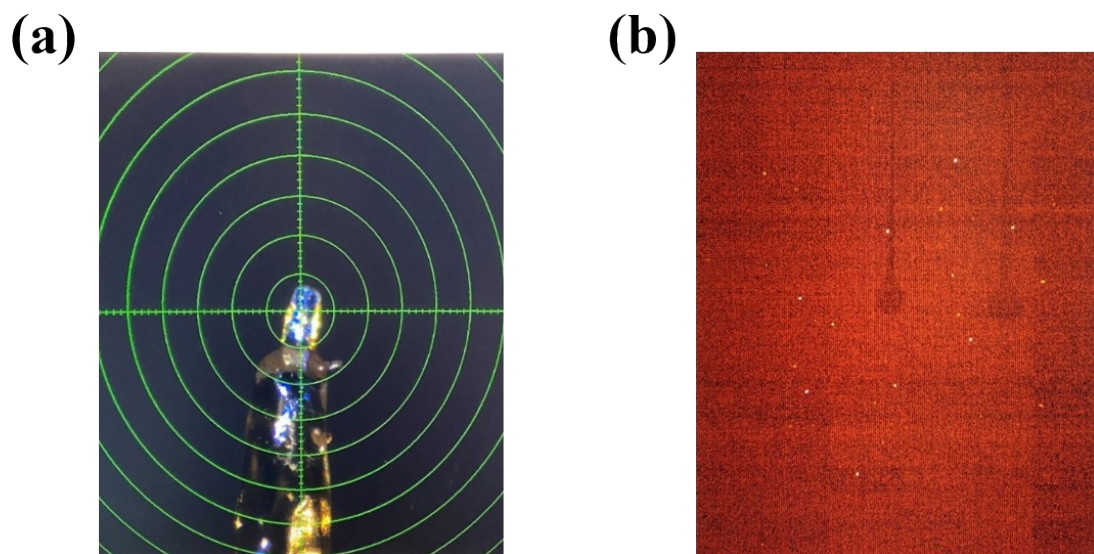


Figure S4. a) Image and b) SCXRD diffraction patterns of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$  single crystal.

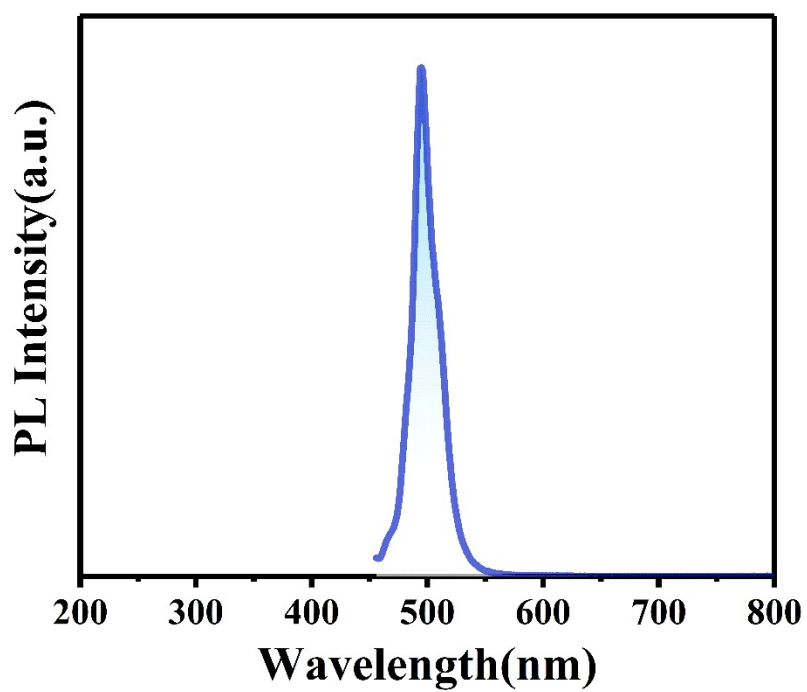


Figure S5. PL spectra of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$ .

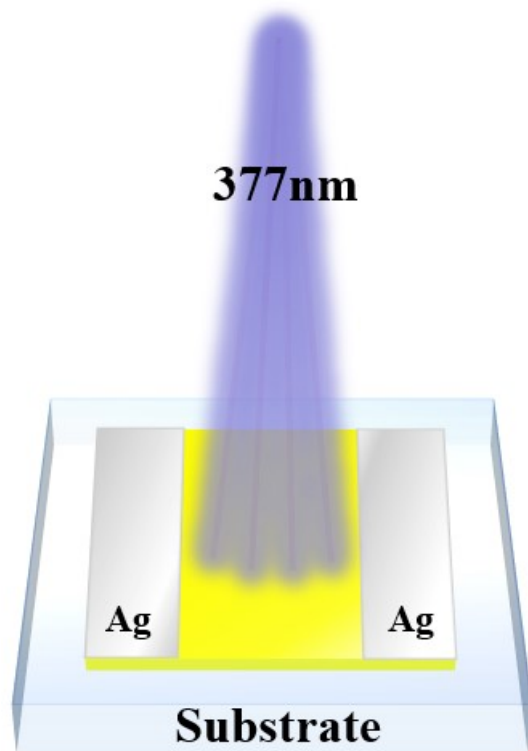


Figure S6. Device structure of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$  crystal-based photodetector.

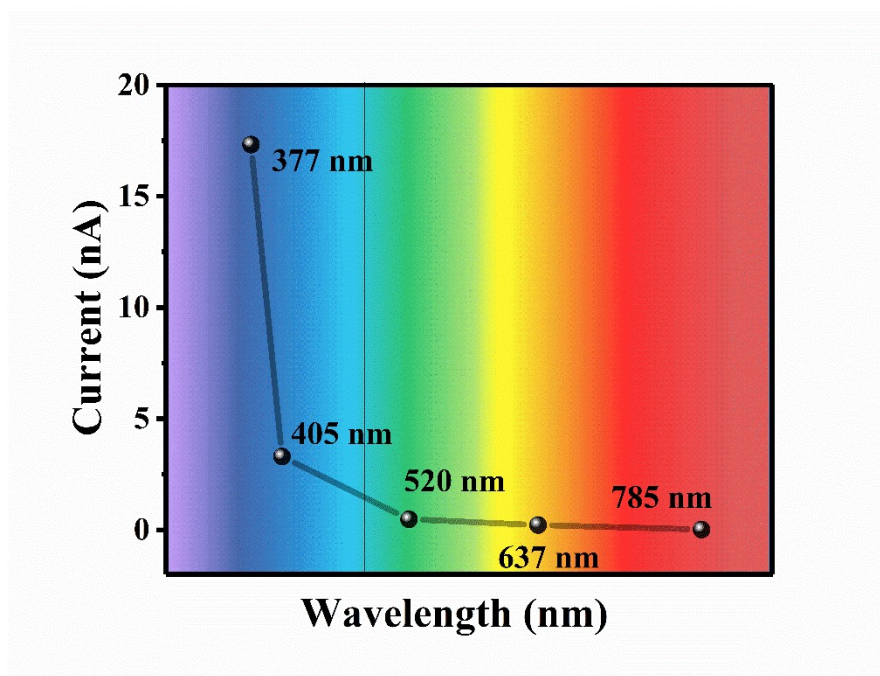
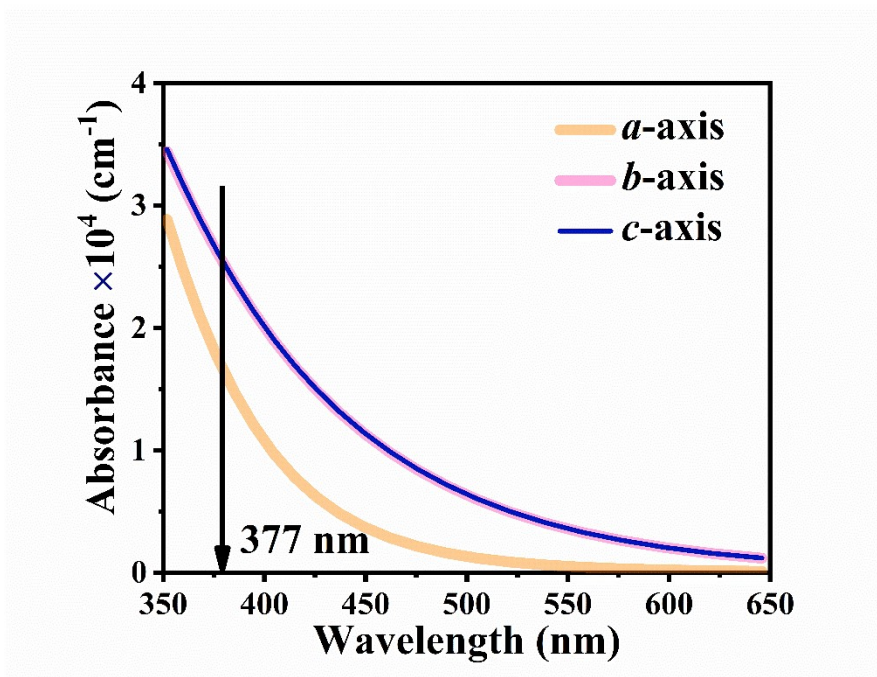
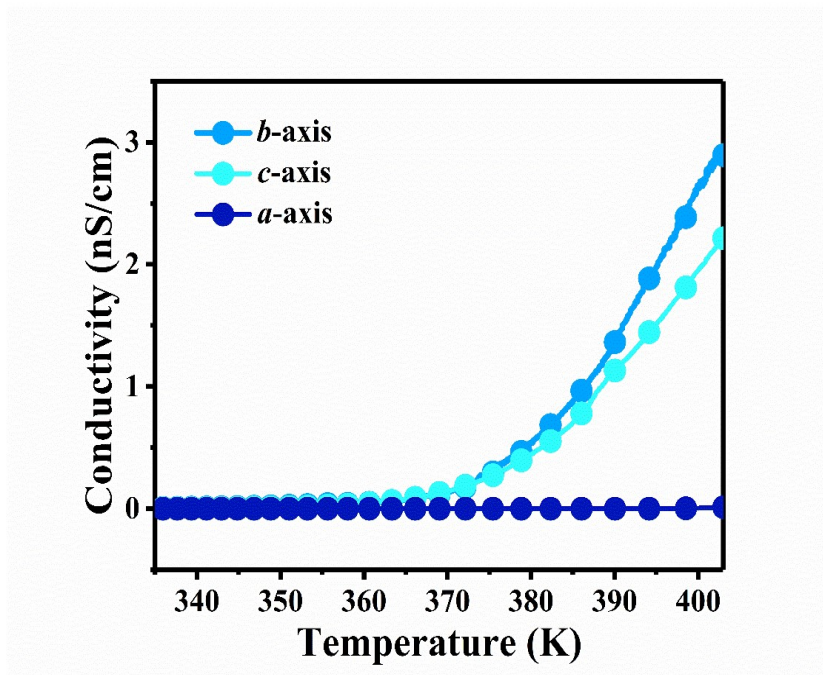


Figure S7. Wavelength-dependent photoresponse of  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$ .

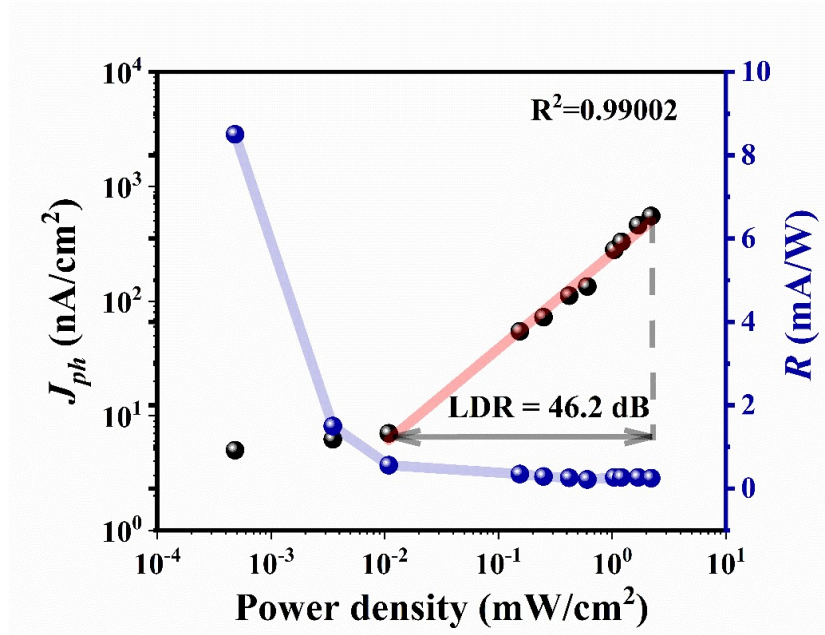


**Figure S8.** Strong anisotropy of optical absorbance along different axes at 377 nm for the crystal of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$ .

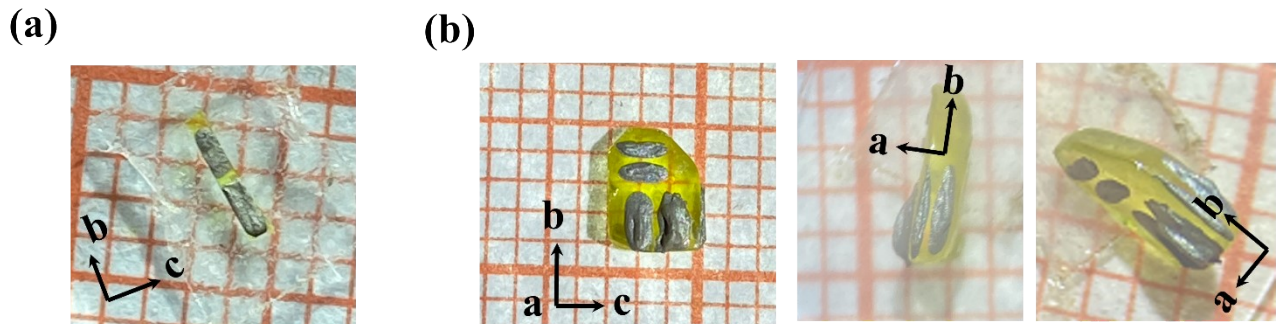


**Figure S9.** Anisotropic conductivity along different axes for the crystal of the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$ .





**Figure S10.**  $J_{ph}$  and  $R$  dependence on the light intensity of the device at the wavelength of 377nm under 10 V bias.



**Figure S11.** (a) The photograph of the planar-type device based on the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$  single crystal for ordinary photodetection tests. (b) The photographs of the planar-type device based on the  $(\text{BA})_2\text{CsPb}_2\text{Br}_7$  single crystal for anisotropic photodetection tests.

**Table S1.** Comparison of anisotropic properties of photodetectors using different materials.

Photodetector	Wavelength (nm)	Bias (V)	Photocurrent anisotropy ratio (Power density)	Ref.
$(\text{C}_6\text{H}_5\text{C}_2\text{H}_4\text{NH}_3)_2\text{PbI}_4$ SC	462	6	57.62 (-)	<b>1</b>
$(\text{NH}_4)_3\text{Bi}_2\text{I}_9$ SC	450	10	$\sim 2$ (0.8 mW/cm <sup>2</sup> )	<b>2</b>

(n-propylammonium)(MA)SbBr <sub>5</sub> SC	405	10	35 (100 mW/cm <sup>2</sup> )	<b>3</b>
(i-BA) <sub>2</sub> CsPb <sub>2</sub> Br <sub>7</sub> SC	425	10	~4 (22.4 mW/cm <sup>2</sup> )	<b>4</b>
(s-BA) <sub>2</sub> (MA)Pb <sub>2</sub> I <sub>7</sub> SC	520	10	~10 (-)	<b>5</b>
(TRA) <sub>2</sub> CsPb <sub>2</sub> Br <sub>7</sub> SC	405	10	~10 (-)	<b>6</b>
(FPEA) <sub>2</sub> PbI <sub>4</sub> SC	520	10	<10 (-)	<b>7</b>
Cs <sub>2</sub> [C(NH <sub>2</sub> ) <sub>3</sub> ]Pb <sub>2</sub> Br <sub>7</sub> SC	405	10	~10 (-)	<b>8</b>
(BA) <sub>2</sub> CsPb <sub>2</sub> Br <sub>7</sub> SC	377	10	25 (26 mW/cm <sup>2</sup> )	<b>This work</b>

i-BA= isobutylamine; s-BA=sec-butylammonium; TRA=(carboxy)cyclohexylmethylammonium; FPEA=p-fluorophenethylammonium.

**Table S2.** Parameter comparison of UV photodetectors using different materials.

Photodetectors	Wavelength (nm)	Bias (V)	Detectivity (Jones)	Response time (rise/decay)	Ref
(BA) <sub>2</sub> CsPb <sub>2</sub> Br <sub>7</sub> SC	377	10	$5.08 \times 10^{11}$	260 $\mu$ s/430 $\mu$ s	<b>This work</b>
MAPbCl <sub>3</sub> SC	365	15	$6.07 \times 10^{11}$	130 ns/365 $\mu$ s	<b>9</b>
EA <sub>4</sub> Pb <sub>3</sub> Cl <sub>10</sub> SC	266	0	$3.06 \times 10^9$	0.8 s/0.22 s	<b>10</b>
MAPbI <sub>3</sub> /TiO <sub>2</sub>	350	1	$2.5 \times 10^{12}$	2 s/1 s	<b>11</b>
SnS <sub>2</sub> /ZnO <sub>1-x</sub> S <sub>x</sub>	365	-	$5.1 \times 10^{10}$	49.51 ms/25.93 ms	<b>12</b>
Few-layers SnS	365	-	$1.92 \times 10^8$	0.3 s/0.5 s	<b>13</b>
[(R)-MPA] <sub>2</sub> PbCl <sub>4</sub> /Si	266	10	$1.2 \times 10^{12}$	500 $\mu$ s/600 $\mu$ s	<b>14</b>
(BPA) <sub>2</sub> PbBr <sub>4</sub>	377	0	~10 <sup>7</sup>	27 $\mu$ s/30 $\mu$ s	<b>15</b>
(PMA) <sub>2</sub> PbCl <sub>4</sub> MMB	320	0	$1.01 \times 10^{11}$	162 $\mu$ s/226 $\mu$ s	<b>16</b>

(R)-MPA=methylphenethylammonium; BPA= 3-bromopropylammonium; PMA= benzylammonium.

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