## **Supporting Information**

## Optical and electronic anisotropies in perovskitoid crystals of

## Cs<sub>3</sub>Bi<sub>2</sub>I<sub>9</sub> studies of nuclear radiation detection

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### **EDS Measurements**

Element	Theoretical atomic	The actual atomic	
	percentage (%)	percentage (%)	
Ι	64.29	64.88	
Cs	21.43	20.14	
Bi	14.28	14.98	
Total:	100.00	100.00	



Fig. SI1 The test spectrum results of EDS of Cs<sub>3</sub>Bi<sub>2</sub>I<sub>9.</sub>



# **Electric Measurements**

Fig. SI2 Typical photoresponse contrast of  $CBI_{(100)}$ ,  $CBI_{(101)}$  and  $CBI_{(001)}$  at 150 V in 425nm LED (~200 mW · cm<sup>-2</sup>).



#### **Radiation detection Characterization**

Fig. SI3 (a), (b) and (c) are the response spectra to <sup>241</sup>Am alpha source of CBI<sub>(001)</sub>, CBI<sub>(101)</sub> and CBI<sub>(100)</sub>, respectively. (d), (e) and (f)  $\mu\tau$  of CBI<sub>(001)</sub>, CBI<sub>(101)</sub> and CBI<sub>(100)</sub> were 7.06×10<sup>-6</sup> cm<sup>2</sup>·V<sup>-1</sup>, 3.90×10<sup>-5</sup> cm<sup>2</sup>·V<sup>-1</sup> and 5.88×10<sup>-5</sup> cm<sup>2</sup>·V<sup>-1</sup> fitted by the single carrier Hecht equation, respectively.



Fig. SI4 (a), (b) and (c) Alpha particles induced pulse shapes of CBI<sub>(001)</sub>, CBI<sub>(101)</sub> and CBI<sub>(100)</sub> under various bias, respectively. (d), (e) and (f) The electron mobilities of CBI<sub>(001)</sub>, CBI<sub>(101)</sub> and CBI<sub>(100)</sub> by the linear fitting, respectively.