Supporting Information

Optical and electronic anisotropies in perovskitoid crystals of
Cs$_3$Bi$_2$I$_9$, studies of nuclear radiation detection

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**EDS Measurements**

Table. SI1 EDS analysis of Cs\textsubscript{3}Bi\textsubscript{2}I\textsubscript{9}.

<table>
<thead>
<tr>
<th>Element</th>
<th>Theoretical atomic percentage (%)</th>
<th>The actual atomic percentage (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>64.29</td>
<td>64.88</td>
</tr>
<tr>
<td>Cs</td>
<td>21.43</td>
<td>20.14</td>
</tr>
<tr>
<td>Bi</td>
<td>14.28</td>
<td>14.98</td>
</tr>
<tr>
<td>Total:</td>
<td>100.00</td>
<td>100.00</td>
</tr>
</tbody>
</table>

Fig. SI1 The test spectrum results of EDS of Cs\textsubscript{3}Bi\textsubscript{2}I\textsubscript{9}.

**Electric Measurements**

Fig. SI2 Typical photoresponse contrast of CBI\textsubscript{(100)}, CBI\textsubscript{(101)} and CBI\textsubscript{(001)} at 150 V in 425nm LED (~200 mW·cm\textsuperscript{-2}).
Radiation detection Characterization

Fig. S13 (a), (b) and (c) are the response spectra to $^{241}$Am alpha source of CBI(001), CBI(101) and CBI(100), respectively. (d), (e) and (f) $\mu\tau$ of CBI(001), CBI(101) and CBI(100) were $7.06\times10^{-6}$ cm$^2$·V$^{-1}$, $3.90\times10^{-5}$ cm$^2$·V$^{-1}$ and $5.88\times10^{-5}$ cm$^2$·V$^{-1}$ fitted by the single carrier Hecht equation, respectively.

Fig. S14 (a), (b) and (c) Alpha particles induced pulse shapes of CBI(001), CBI(101) and CBI(100) under various bias, respectively. (d), (e) and (f) The electron mobilities of CBI(001), CBI(101) and CBI(100) by the linear fitting, respectively.