

*Electronic Supporting Information (ESI)*

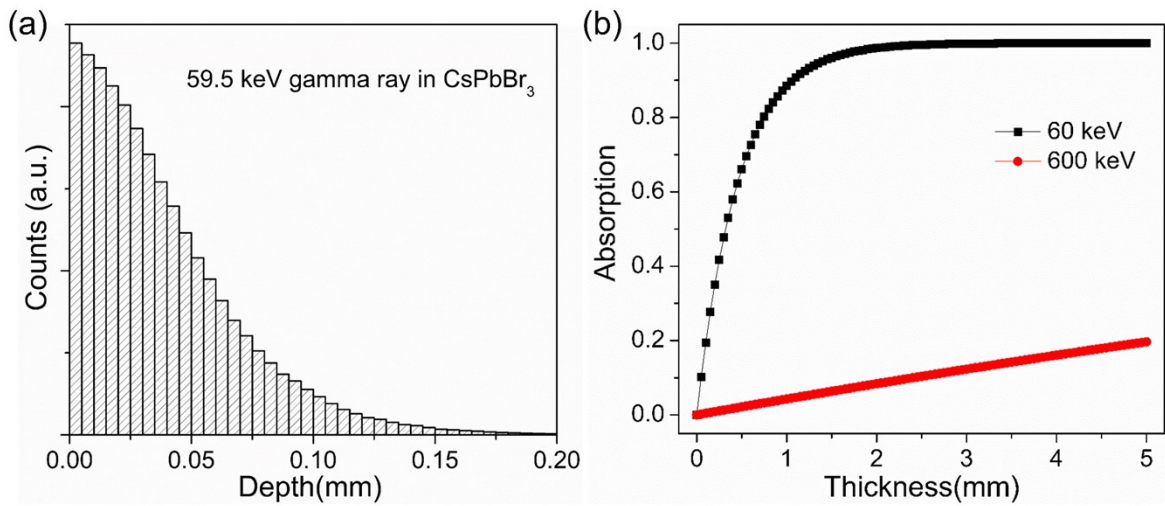
**Investigation on Energy Resolution of CsPbBr<sub>3</sub> Detectors: from Charge Transport Behaviors to Device Configuration**

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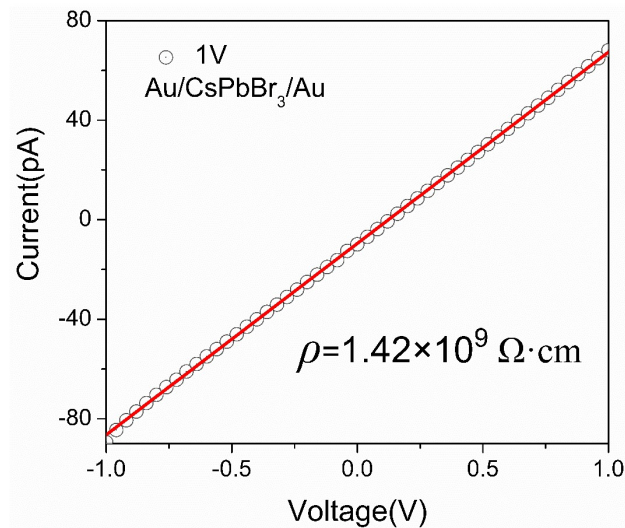
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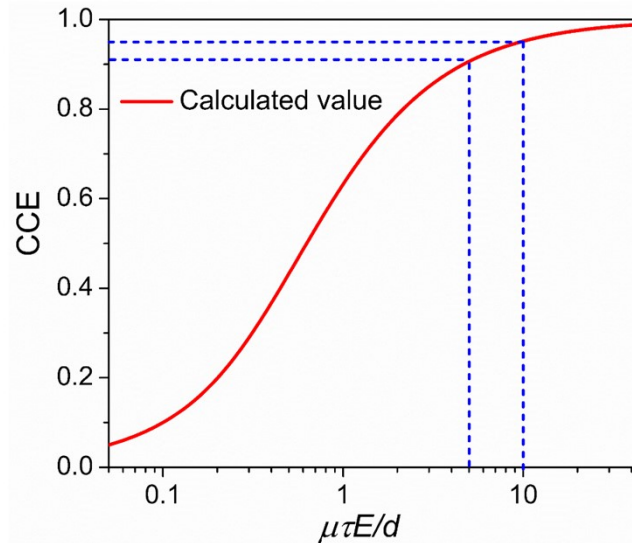


**Figure S1.** (a) Simulation results of incident depth of 59.5 keV  $\gamma$ -ray in CsPbBr<sub>3</sub>. (b) Absorption rate of different energies of  $\gamma$ -rays in CsPbBr<sub>3</sub> crystals with different thicknesses.

According to the energy absorption efficiency spectra as shown in Figure S1a, the <sup>241</sup>Am 59.5 keV  $\gamma$ -ray could be completely absorbed by CsPbBr<sub>3</sub> single crystal of 1.5 mm thickness. The 2-mm-thick CsPbBr<sub>3</sub> used in simulation work attenuated almost all the energy of the 59.5 keV  $\gamma$ -ray source. For  $\gamma$ -ray above 600 keV, the radiation can be treated as evenly deposited within the 5 mm CsPbBr<sub>3</sub> crystal (Figure S1b).



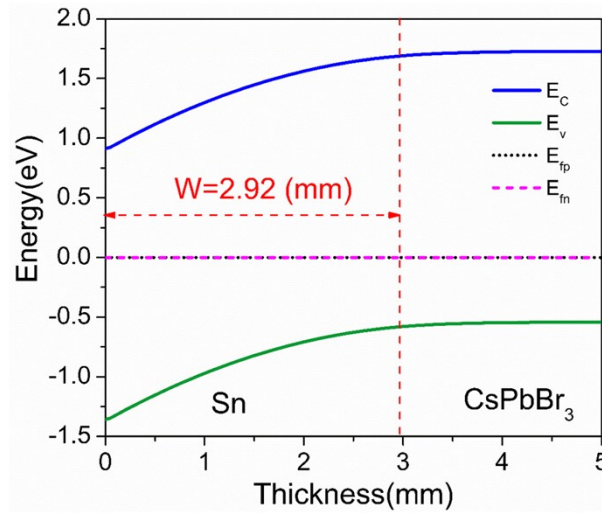
**Figure S2.** The typical  $I$ - $V$  curve of the CsPbBr<sub>3</sub> crystal



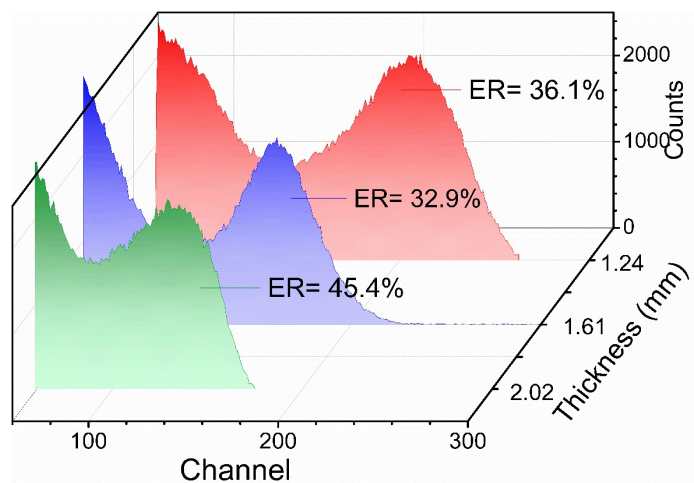
**Figure S3.** The dependence of CCE on  $\mu\tau E/d$

When the detector is illuminated from the anode, the single charge carrier approximation Hecht equation is:

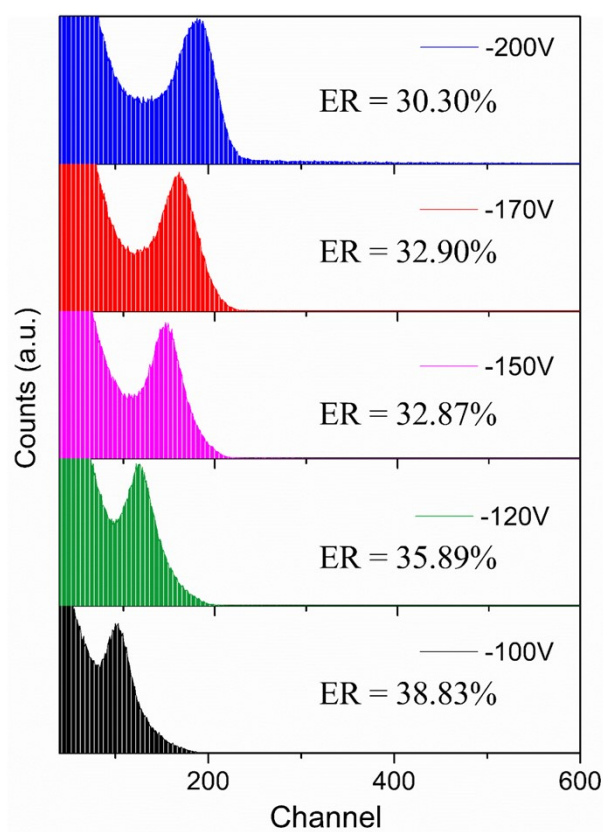
$$CCE = \frac{(\mu\tau)_h E}{d} \left( 1 - e^{-\frac{d}{(\mu\tau)_h E}} \right) \quad (S1)$$



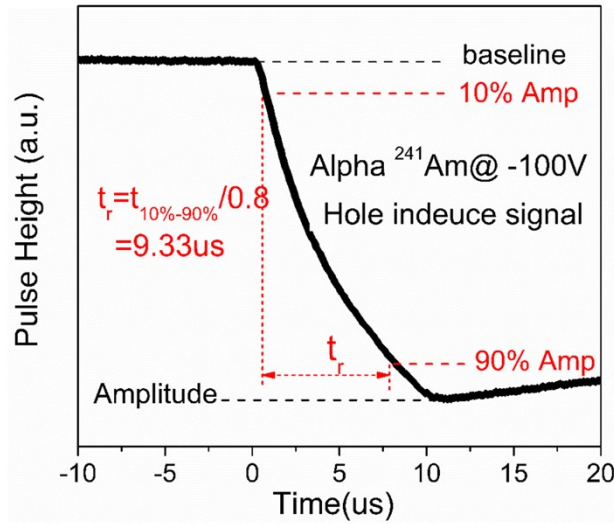
**Figure S4.** Band diagram of Schottky contact



**Figure S5.**  $^{241}\text{Am}$   $\gamma$ -ray spectra acquired by the planar detector with different thicknesses.

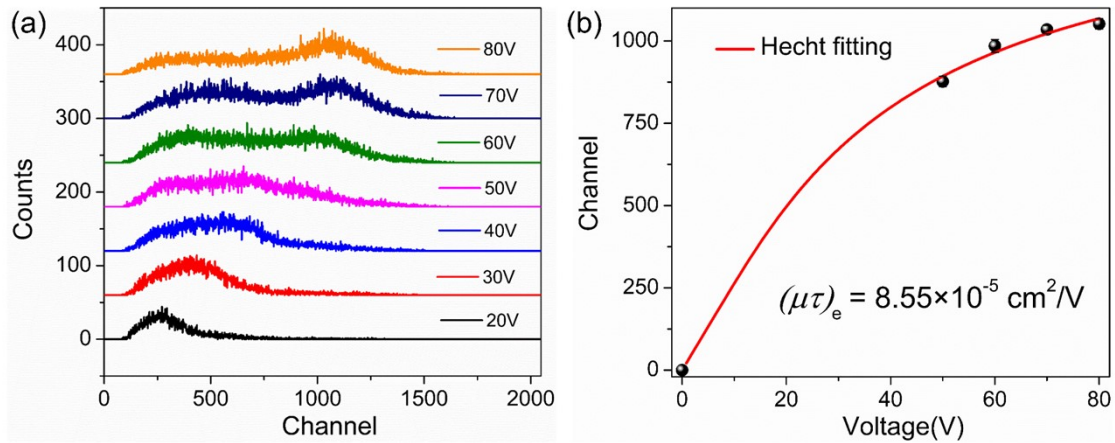


**Figure S6.**  $^{241}\text{Am}$   $\gamma$ -ray spectra acquired by the planar detector with different applied voltages.

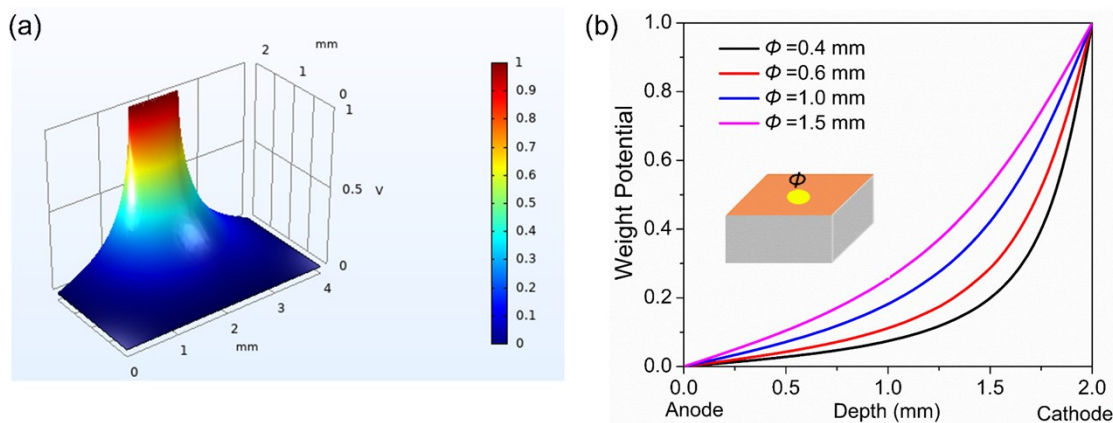


**Figure S7.** Typical hole transport voltage pulse of Sn/CsPbBr<sub>3</sub>/Au device irradiated by anode of  $\alpha$ -particles under -100 V bias

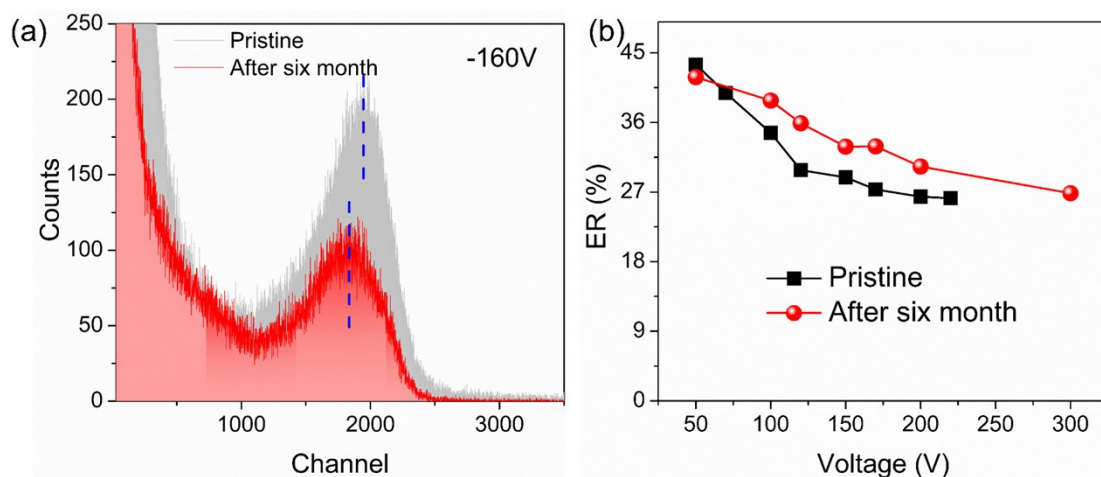
Figure S7 shows a typical transient pulse from the CsPbBr<sub>3</sub> device induced by <sup>241</sup>Am  $\alpha$ -particles incident on the anode at a bias of -100 V, produced in this case by the transport of holes across the active thickness of the device. By collecting the fall time of 1000 pulses under various bias voltages, the average drift time ( $t_{dr}$ ) of holes can be obtained



**Figure S8.** (a)  $\alpha$ -particle energy spectra of electron at different voltages. (b) The electron mobility lifetime product fitting by Hecht equation.



**Figure S9.** (a) Simulated weighted potential of the cross section in the quasi-hemispherical device. (b) Simulate the quasi-hemispheric weight potential distribution with different anode radius.



**Figure S10.** (a) The  $^{241}\text{Am}$  @5.5 MeV  $\alpha$ -particle measurements with the as-grown crystal and the crystal after six months. (b) The ER of newly grown and 6-month-old planar detectors for  $^{241}\text{Am}$  59.5-keV at different voltages.

Bear in mind, the device is placed in the air without water and oxygen isolation package, so the water vapor in the air will have a slight impact on the performance of the device, resulting in a slight deterioration of the ER. But in general, the change of ER within six months is not significant.

**Table S1.**  $\alpha$ -particle measurements data will deteriorate due to surface aging.

	Pristine	After six month
$(\mu\tau)_h$ ( $\text{cm}^2 \text{V}^{-1}$ )	$1.77 \times 10^{-4}$	$1.01 \times 10^{-4}$
$\mu_h$ ( $\text{cm}^2 \text{V}^{-1} \cdot \text{s}^{-1}$ )	47	43.4
$\tau_h$ ( $\mu\text{s}$ )	3.76	2.32