High performance detection for x-ray and γ-ray with $MAPbX_3$ perovskite single crystals

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Recently, organometallic lead trihalide perovskites, such as $MAPbBr_3$ (MA = Methylammonium, Pb = Lead, Br = Bromide), have emerged as a new generation of opto-electronic materials[1]. Except for the applications in solar cells[2,3] and light-emitting diodes[4], perovksites have also been widely used for x-ray and γ-ray detections due to their high-Z elements (Pb, Iodide, and Br) with respect to the high energy photons stopping power (or attenuation coefficient), low trap-state density, long carrier diffusion, and tunable energy bandgaps[5-7]. Comparing with the commercial x-ray and γ-ray detectors, i.e. α-Se detector and CdZnTe detector, the high performance detection for x-ray and γ-ray with $MAPbX_3$ is still a big challenge.

For x-ray and γ-ray detections, the detectors should have high sensitivity. If the photon counting method is adopted, the high energy resolution and high time resolution are also required. According to these requirements, the high performance detector for x-ray and γ-ray with $MAPbX_3$ must have large quantum efficiency, long carrier diffusion length, small noises, and very short temporal response time. In this work, the large area $MAPbBr_3$ single crystal has been fabricated with a facile methodology. Due to the quite thick active material and large carrier mobility, the x-ray photons and γ-ray photons can be absorbed with high efficiency. The photo generated electrons and holes can also be collected effectively with the large electric field. To decrease the dark current in the detection, a novel photo-diode structure is proposed here. In crystallization process of $MAPbI_3$ single crystal, the p-n junction can be formed with doping of selenium atoms into $MAPbI_3$ single crystal.

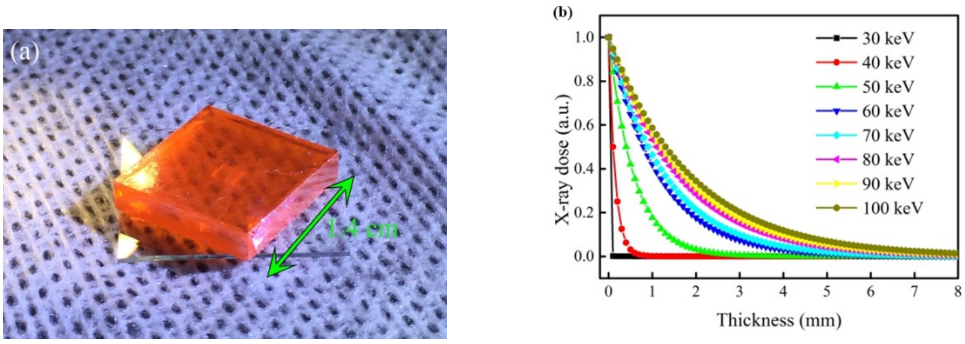

Fig. 1(a) photo of $MAPbBr_3$ single crystal grown by various temperature crystallization; (b) Absorption of x-ray with different thickness of $MAPbBr_3$ single crystal.

Fig.1(a) is the optic photo of large $MAPbBr_3$ single perovskite crystal ($MAPbBr_3$ SPC 30mm×30mm×7mm) fabricated with various temperature method. The absorption of x-ray does with different thickness of $MAPbBr_3$ SPC is shown in Fig.(b). It can be seen that almost all of the x-ray photons are absorbed when the $MAPbBr_3$ SPC is 7mm thick. The experimental results also shown the sensitivity is as high as $305\mu C\ Gy_{air}^{-1}cm^{-2}$ when the anode voltage of x-ray tube is 30kV.

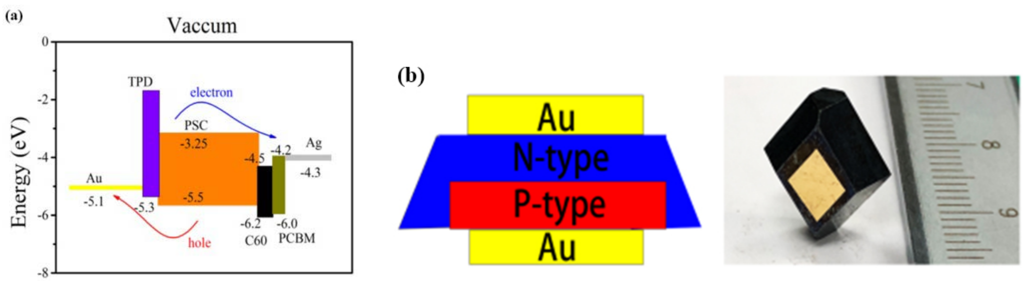

Fig. 2 (a) Energy diagram of multi layers PSC photo diode; (b) PSC photo diode developed by the doping of Se atoms in the crystallization process of $MAPbI_3$

To reduce the dark current in the detection, two type of photo diode structures have been proposed here. In Fig.2(a), a photo diode with structure of Au/TPD/$MAPbBr_3$ PSC/C_{60}/PCBM/Ag has been fabricated with spin coating and sputtering methods. Although the dark current density can be reduced to 20 nA/cm^2 with -30V bias voltage, the temporal response time is nearly 50 μs due to the defects on the interfaces between PSC and carriers transport layers. In Fig.2(b), by doping selenium (Se) in $MAPbI_3$ perovskite single crystals (DPC) crystallization process, low dark current p-n junctions were fabricated without any organic layers. After two golden electrodes deposited on opposite faces of $MAPbI_3$ DPCs, high performance photodiodes can be achieved.

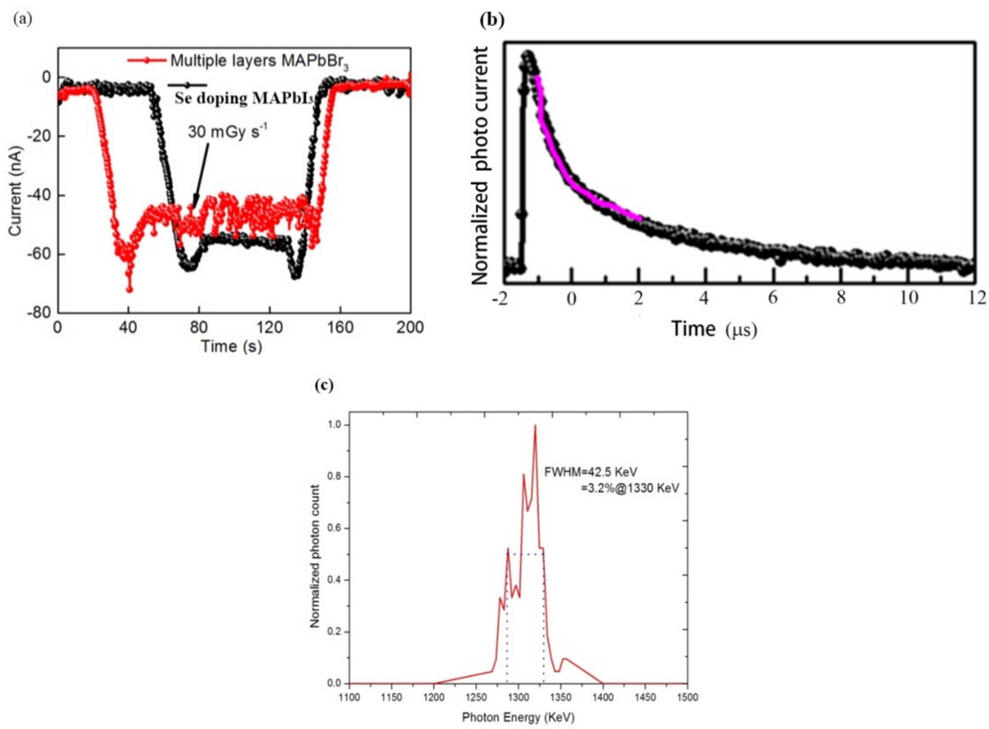

Fig. 3 (a) Photo current illuminated by γ photons of 1.33 MeV; (b) Temporal response time of $MAPbI_3$ DPC; (c) Energy resolution of γ-ray detection with $MAPbI_3$ DPC

As the experimental results shown, the photodiodes shown in Fig.2(b) gives the high detection sensitivity as 21000 μC $Gy_{air}^{-1}cm^{-2}$ and 41 μC $Gy_{air}^{-1}cm^{-2}$ for 60 keV x-ray and 1.33 MeV γ-ray respectively. In this photodiode, the transition time becomes shorter under higher electric field, and the carrier lifetime also becomes shorter due to the dopant of Se atoms. Finally, the temporal response time is measured as 3 μs by experiments. The FWHM width of energy spectrum is decreased to 3.2%@1330 keV.