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Long-Term High-Voltage Bias Stability and Degradation Pathways in Perovskite Single-Photon Counting Radiation Detectors

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Current existing semiconductor radiation detection technologies, such as Cadmium Telluride (CdTe), Cadmium Zinc Telluride (CZT), High-purity Germanium (HpGe) and Silicon (Si), suffer from several limitations. These include high production costs, the need for cryogenic cooling (for HpGe), and insufficient stopping power in materials composed of low-atomic-number elements (such as Si). These challenges have motivated the radiation detection community to investigate novel materials that offer lower production costs, the ability to operate at room temperature, and high stopping power for semiconductor radiation detection technologies.

Perovskite Single Crystals (PSCs) have emerged as a promising candidate for direct radiation detection in photon-counting mode due to their excellent optoelectronic properties, low defect density, and high mobility-lifetime products (μ⊠). Despite these advantages, ion migration in PSCs poses a significant obstacle for the stable long-term operation of these devices. Under high electric fields, ion migration accelerates the electrochemical reactions at the metal/perovskite interface, leading to device degradation. This remains a significant barrier to the commercialisation of PSC-based radiation detectors.

In this work, hybrid PSCs (FAPbBr3) are grown from solution using a low-cost, low-temperature method called inverse temperature crystallisation (ITC). These PSCs are polished to achieve smooth, mirror-like surfaces, and metal electrodes, such as Bi and Au, are deposited by thermal evaporation. Both single-pad and guard-ring device architectures are fabricated. These detectors exhibit excellent charge-transport properties, including high hole mobilities of 190 cmˆ2 Vˆ(-1) sˆ(-1) and a high hole μ⊠ of 2.7 x 10 ˆ(-3) cmˆ2 Vˆ(-1). Importantly, guard ring devices demonstrate an order of magnitude reduction of dark current –from 29 nA cmˆ(-2) in single pad detectors to 2.9 nA cmˆ(-2) in guard ring devices, highlighting the importance of electrode engineering on suppressing the leakage currents of these devices.

Long-term dark current measurements were conducted for both device types, and X-ray photoelectron spectroscopy (XPS) was performed to characterise the chemical composition and bonding environment of the crystal's surface and bulk following extended device biasing. Additionally, the implementation of charge-blocking layers via atomic-layer deposition (ALD) is explored as a strategy to mitigate ion migration toward the anode metal electrode. Finally, long-term single-photon-counting radiation-detection measurements are performed using 241Am 5.49 MeV α-particles, 59.5 keV γ-rays, and 137Cs 662 keV γ-rays to investigate the device's long-term operation under constant radiation exposure.

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