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## On the Detection of High-Energy Protons by Perovskite Single Crystals: Enhanced Device Performance and Healing in MAPbBr3 and MAPbI3 Single Crystal-based Systems after Exposure to Protons

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Proton detectors play a crucial role in various fields, including radiation monitoring, medical imaging, and particle physics research. Recently, metal halide perovskites have emerged as a promising candidate for proton detection, offering high radiation tolerance alongside their demonstrated remarkable capabilities in detecting X-rays, gamma rays, alpha particles, electrons, and neutrons. These materials exhibit excellent charge transport characteristics, fast response times, high stopping power, and cost-effectiveness. Their versatile nature allows them to effectively capture and measure radiation across a wide spectrum, making them valuable for diverse radiation detection and imaging applications.

In this study, we fabricate proton detectors based on inverse temperature crystallization (ITC) grown methylammonium lead tribromide (MAPbBr3) and methylammonium lead triiodide (MAPbI3) single crystals using a coplanar structure. We then measure their response under high energy proton irradiation (68 MeV), commonly used for proton therapy of eye tumors, and confirm their ability to detect protons. Additionally, we investigate their stability up to a total proton dose of 10<sup>13</sup> p/cm<sup>2</sup>. Interestingly, we observe an initial degradation followed by a healing process that improves sensitivity by 6-fold and 202-fold for MAPbBr3 and MAPbI3-based detectors, respectively.

To confirm this healing effect, we then measure and evaluate transient photoluminescence (TRPL), photoresponse, and external quantum efficiency (EQE) after proton irradiation. We observe an increase in signalto-noise (S/N) ratio by a factor of 11 for MAPbBr3 and 5 for MAPbI3-based detectors in photoresponse corroborating the improved detection capabilities, while longer TRPL lifetimes suggest improved charge carrier dynamics within the detector material. Moreover, we observe higher EQE indicating an improved efficiency in converting incident radiation into electrical signals. Together, these intriguing findings suggest a protoninduced self-healing of the perovskite proton detectors leading to enhanced functionality and improved performance. The results hence, highlight the suitability of MAPbBr3 and MAPbI3-based detectors for proton detection. Overall, these findings contribute to the ongoing efforts in advancing proton detection technology and pave the way for more accurate and reliable proton-related research and applications in various fields.

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