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Optical characteristics and scintillation processes in 2D perovskite radiation detectors

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This study investigates the fundamental performance of perovskite scintillators for X-ray and gamma imaging, with a particular focus on their potential applications in nuclear security detection. By combining unique structural and optoelectronic properties, these materials offer promising prospects for developing next-generation radiation detection technologies, aligning with the goal to enhance detection capabilities for nuclear security.

Two-dimensional (2D) hybrid perovskites represent a versatile class of materials that combine the structural diversity of organic molecules with the ideal optoelectronic properties of inorganic frameworks. In this study, we present a comprehensive investigation of the optical and scintillation processes in 2D perovskites with Ruddlesden-Popper (RP) and Dion-Jacobson (DJ) structures. The materials studied include BA2PbBr4, PEA2PbBr4, BA2PbI4 (all RP), and TMPDAPbBr4 (DJ). Through steady-state and time-resolved photoluminescence (PL), X-ray and alpha scintillation measurements, and structural analysis, we show the correlation between their structural motifs and optoelectronic performance.

The bromide-based RP perovskites, BA2PbBr4 and PEA2PbBr4, exhibit bright photoluminescence, sharp emission spectra, and relatively small Stokes shifts. This is likely due to their efficient charge-carrier dynamics, with minimal non-radiative losses. These materials also show promising alpha particle scintillation responses, reinforcing their potential for radiation detection applications. However, the iodide-based BA2PbI4 displays dimmer photoluminescence and lower scintillation yields, potentially due to its small negative Stokes shift, which increases the likelihood of reabsorption of emitted photons. This effect, combined with the potential for enhanced non-radiative recombination pathways in the iodide system, likely contributes to the observed reduction in optical and scintillation performance. These three RP perovskites have very fast decay times in the several nanoseconds, making them ideal for a variety of nuclear security applications.

In contrast, the DJ perovskite TMPDAPbBr4 demonstrates distinctly different behaviour to the RP perovskites, characterised by a broad PL emission profile and a significantly larger Stokes shift. Just like the RP perovskites, TMPDAPbBr4 also has a fast decay time which is unusual for broad emitters. These features suggest the involvement of additional recombination mechanisms, likely mediated by defects or self-trapped excitonic states. Despite its rapid decay dynamics, TMPDAPbBr4 retains sufficient radiative efficiency to enable measurable scintillation, albeit with different characteristics compared to the RP materials.

The findings of this study highlight the influence of compositional and structural tuning on the optical and scintillation properties of 2D perovskites. While RP perovskites exhibit more conventional excitonic behaviour with narrowband emission, the DJ perovskite shows the potential for broader and more complex emission pathways. These insights provide a foundation for advancing the design of 2D perovskites tailored for applications in scintillation and radiation detection, where both high efficiency and fast response are critical.

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