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Bulk and interfacial ion dynamics in quasi-2D and 3D perovskite LEDs

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The optimal performance of both photovoltaic and electroluminescent devices necessitates minimal nonradiative recombination losses. In perovskites, such losses are significantly influenced by carrier traps associated with mobile ions and vacancies, resulting in I–V hysteresis in solar cells and affecting the performance of perovskite-based optoelectronic devices, including photodetectors and LEDs.[1][2] Ideally in a perovskite LED (PeLED) operation, there are cations at the hole injection layer (HIL)/perovskite interface and anions at the electron injection layer (EIL)/perovskite interface, and upon the application of a forward bias, there is a redistribution of the ions at opposite interfaces under sufficient time scales. This consequently alters the energy landscape in a manner that promotes increased radiative recombination in the bulk.[3] In this work, we study ion dynamics at both the HIL and EIL interfaces along with the bulk of quasi-2D and 3D PeLEDs with time-resolved electroluminescence (TrEL) measurements, where we observed that with shorter pulse periods and longer pulse durations, the electroluminescence of quasi-2D PeLEDs underwent significant degradation compared to their 3D counterpart, which showed an opposite trend under similar conditions. We attribute this to the screening of the field at the HIL and EIL interfaces in the quasi-2D PeLEDs due to the presence of bulky spacer cations of the 2D perovskite lattice, which hamper the ionic redistribution at interfaces [4], thus mitigating the radiative recombination in the bulk. The presence of a large number of grain boundaries in quasi-2D PeLEDs is another factor contributing to strong ionic migration pathways under pulsed operations in their bulk. [5] In this work we corroborate the TrEL results with cross-section Kelvin probe force microscopy (KPFM) analysis and conductive atomic force microscopy (cAFM) studies performed under similar pulsed operations to study the ionic dynamics at the interface and the bulk. We also plan to investigate qualitatively the ionic species, mainly uncoordinated lead (Pb_i) and lead vacancies (V_{Pb}) formed under such pulsed operations through Deep Level Transient Spectroscopy (DLTS) under similar pulsed conditions implemented in TrEL measurements. Finally, to mitigate these field-induced traps, targeted bulk and interface passivation strategies will be deployed, incorporating novel charged dopants to passivate ionic species, suppress non-radiative recombination centers, and stabilize the quasi-2D PeLED lattice.

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