

# Stabilizing perovskite light-emitting diodes by incorporation of binary alkali cations

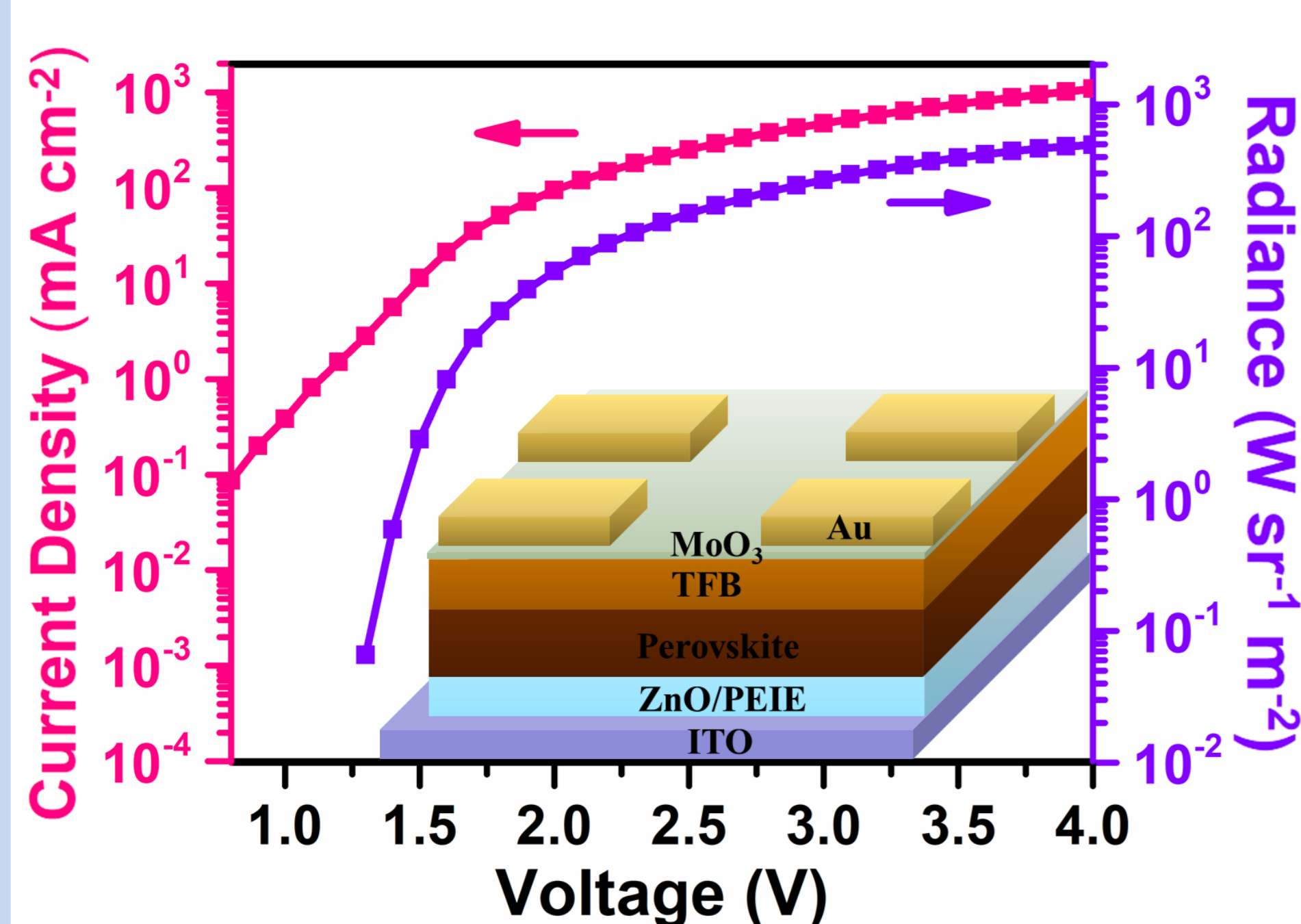
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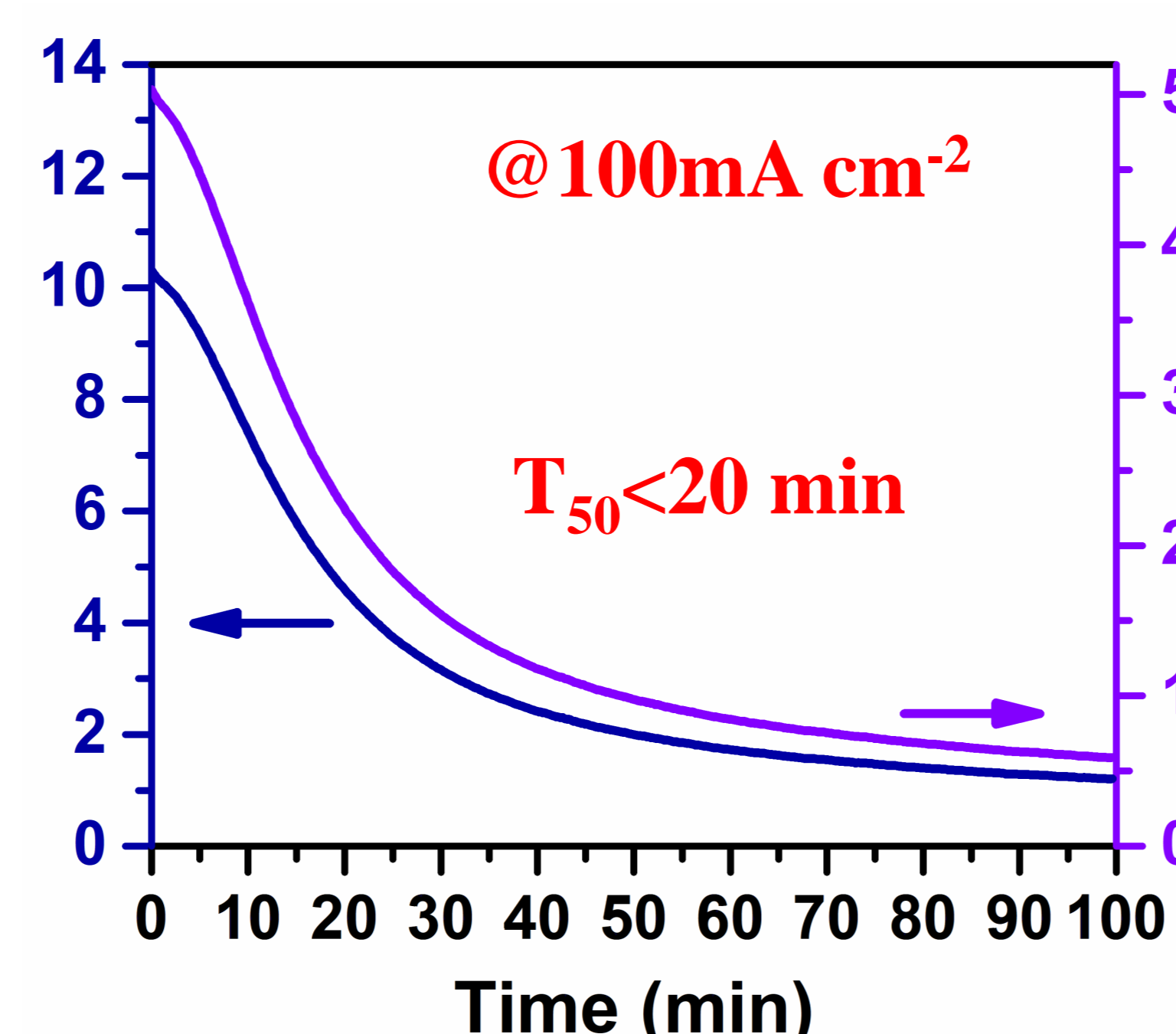
## Abstract

Poor stability of perovskite light emitting diodes (PeLEDs) is a key bottleneck that hinders commercialization of this technology. In our recent work, using time-of-flight secondary ion mass spectrometry we find the degradation of FAPbI<sub>3</sub>-based PeLEDs during operation is directly associated with ion migration and incorporation of binary alkali cations, i.e., Cs<sup>+</sup> and Rb<sup>+</sup>, in FAPbI<sub>3</sub> could suppress ion migration and significantly enhance the lifetime of PeLEDs. Combining experimental and theoretical approaches, we further reveal that Cs<sup>+</sup> and Rb<sup>+</sup> ions stabilize the perovskite films by locating at different lattice positions, with Cs<sup>+</sup> ions present relatively uniformly throughout the bulk perovskite while Rb<sup>+</sup> ions preferentially on the surface and grain boundaries. Further chemical bonding analysis show that both Cs<sup>+</sup> and Rb<sup>+</sup> ions raise net atomic charges of surrounding I anions, leading to stronger Coulomb interactions between the cations and the inorganic framework. The PeLEDs show significantly enhanced operation stability, achieving a half lifetime over 3600 min.

## Stability issue of PeLEDs under operation

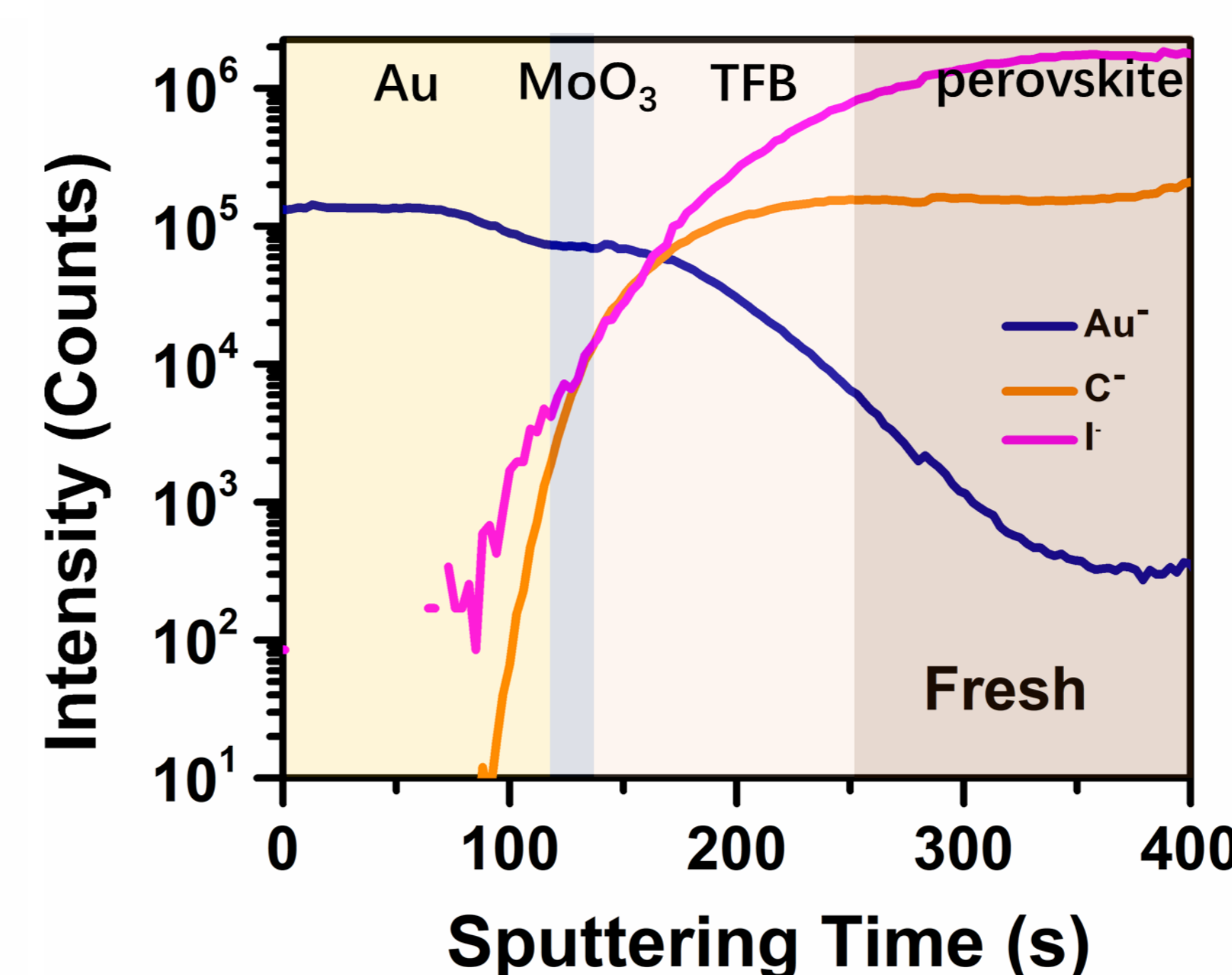


FAPbI<sub>3</sub>-based PeLEDs



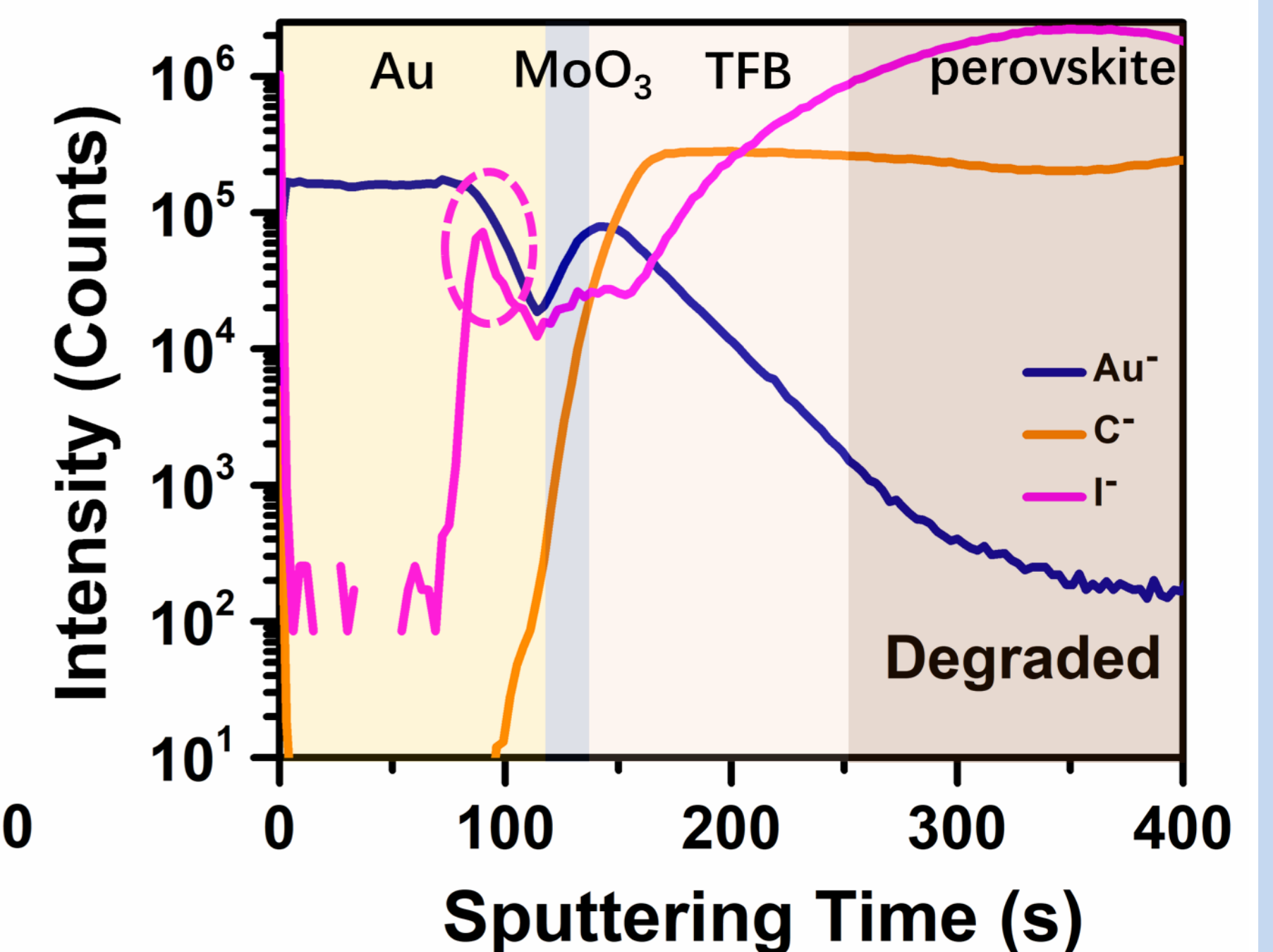
Device degradation under a constant current density

## Ion-migration under bias

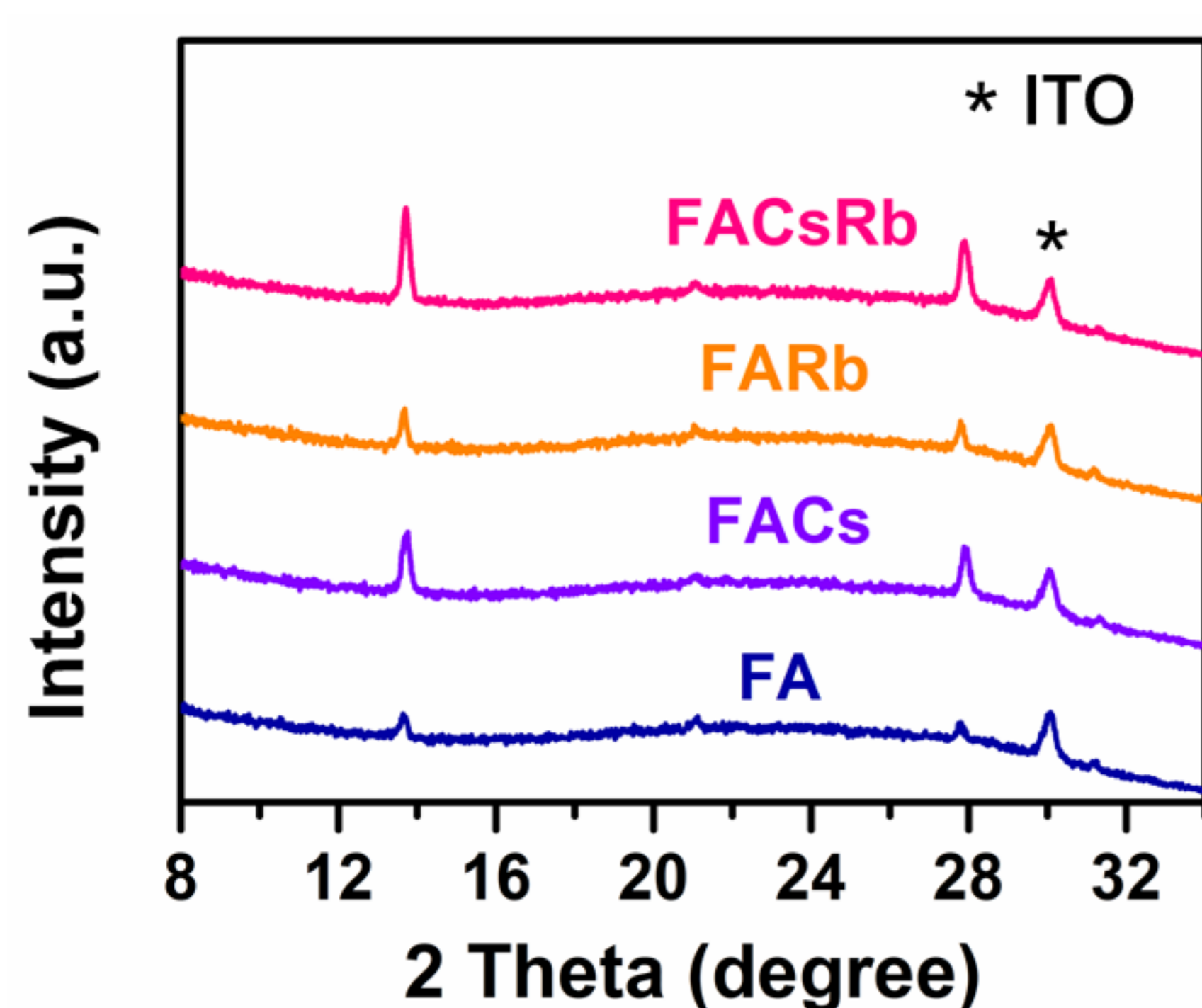


Distribution of ion species in the fresh (left) and degraded (right) device measured by TOF-SIMS depth profiling

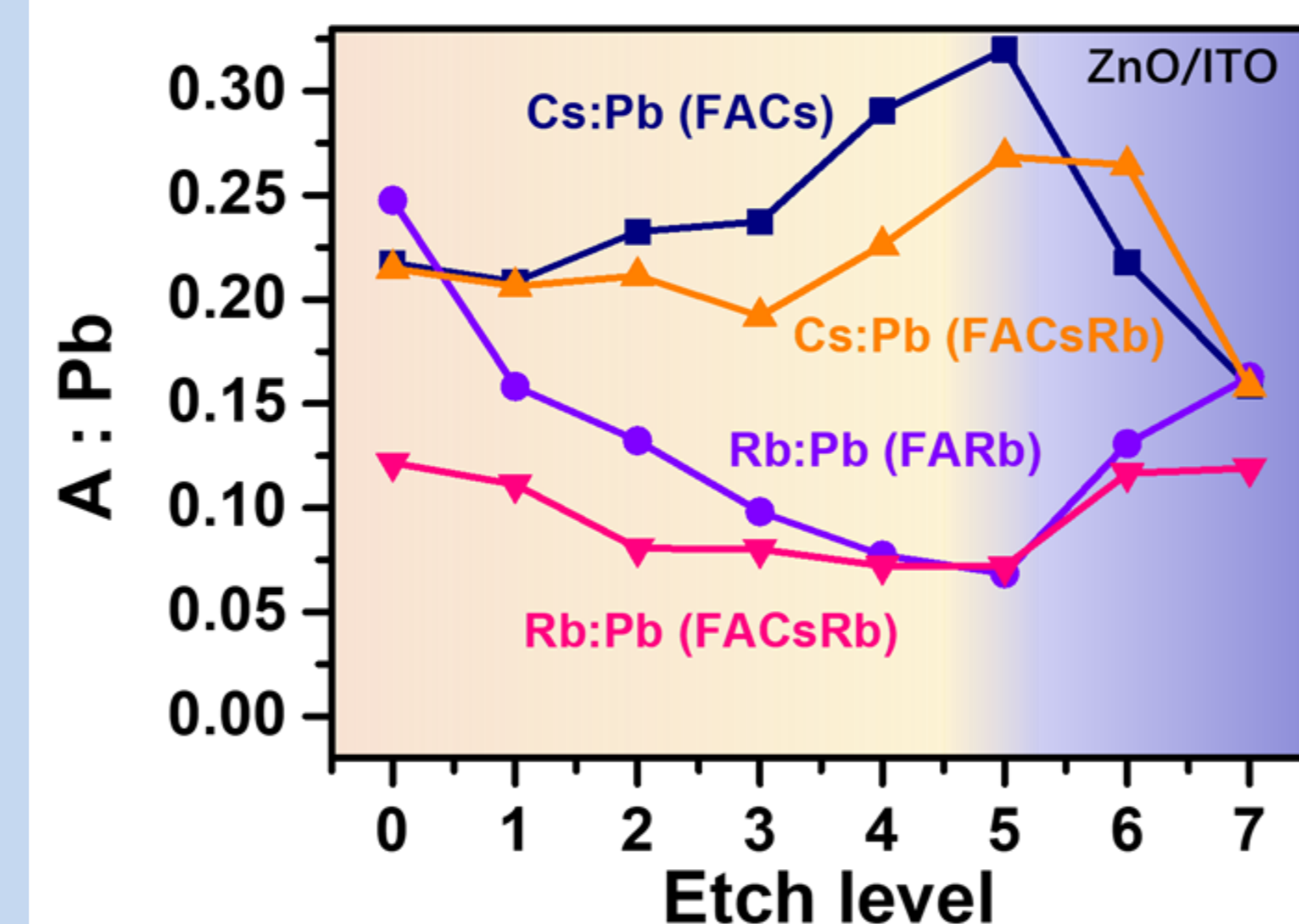
1. defects-assisted
2. main migration of halide ions



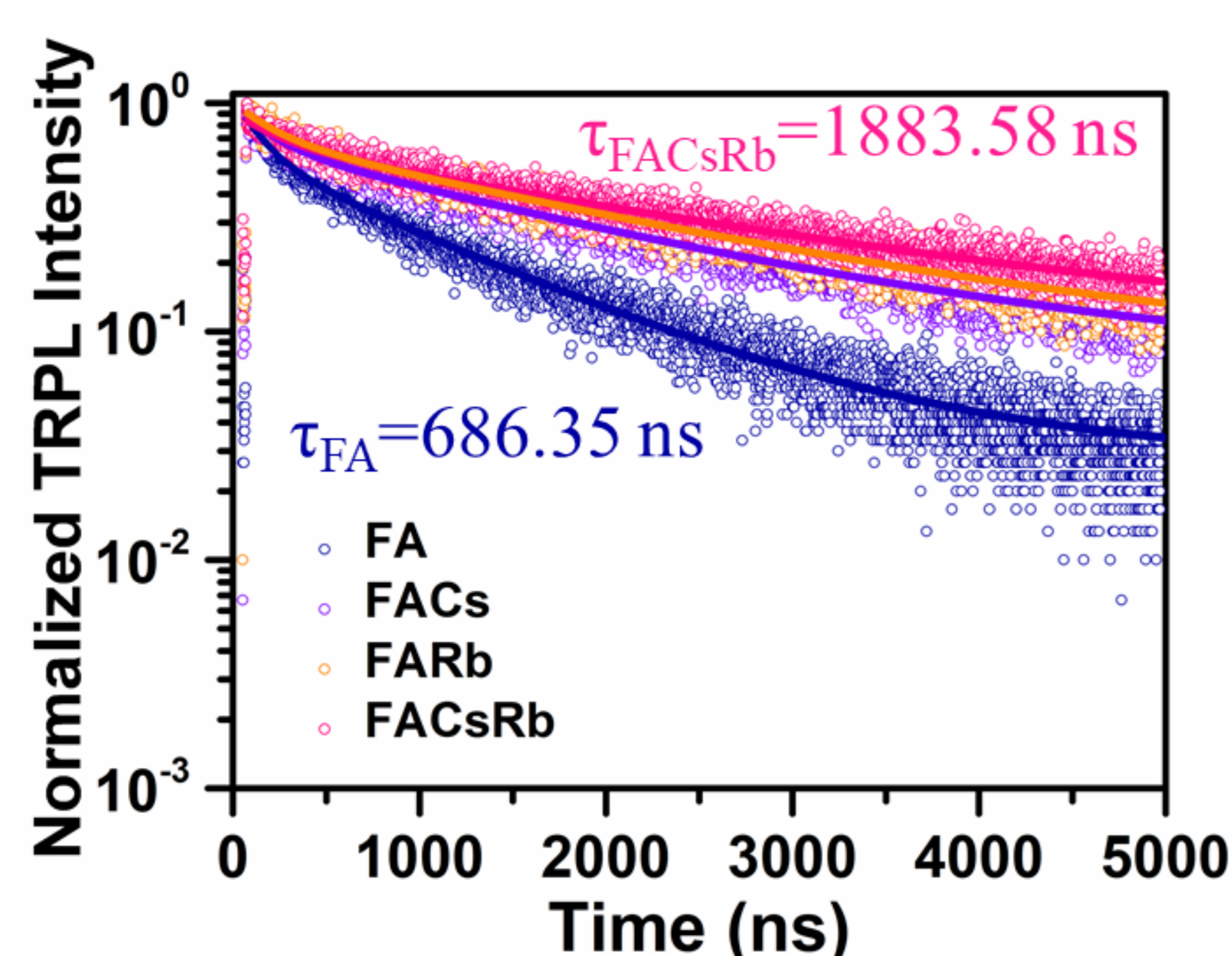
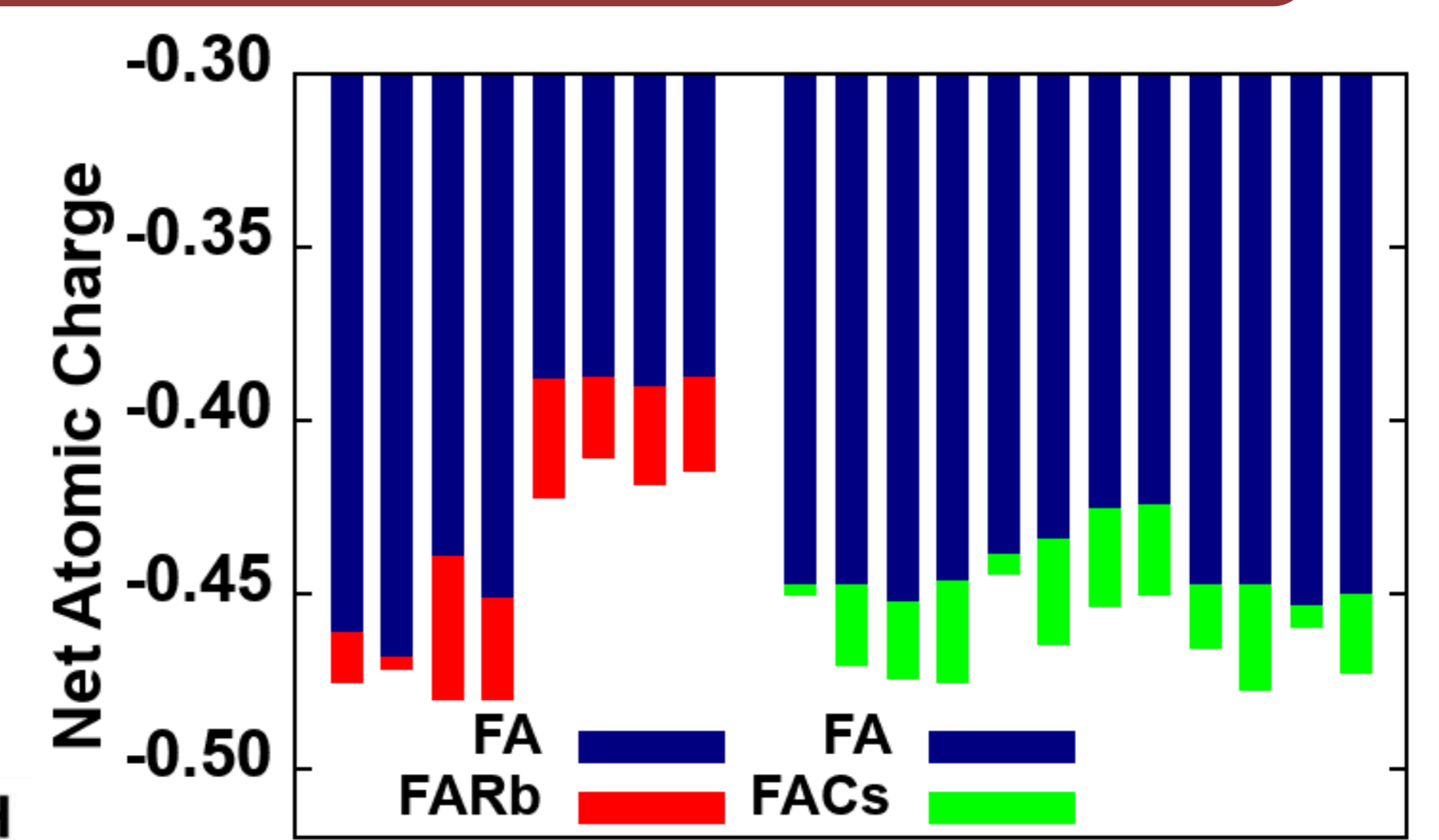
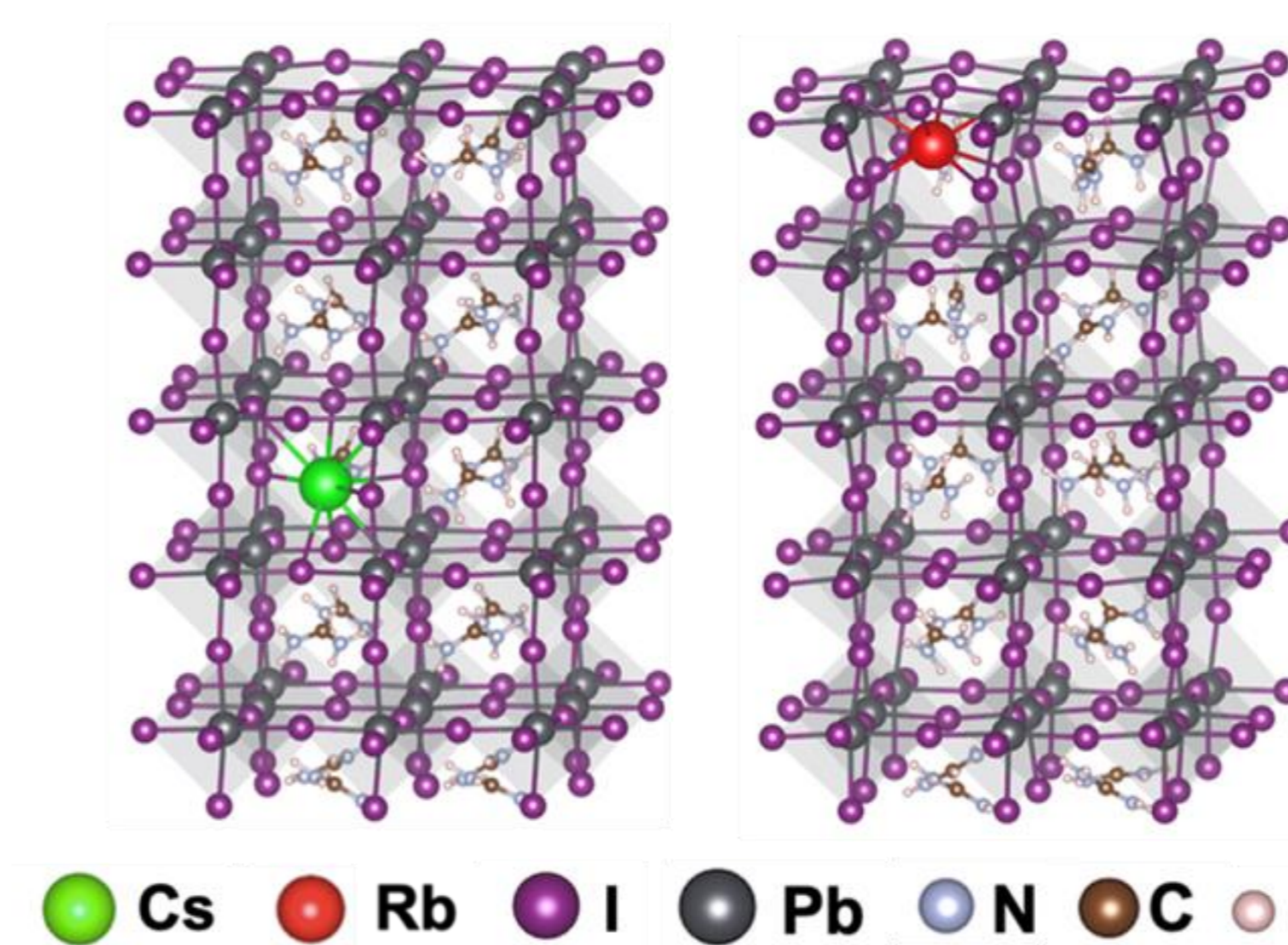
## Films doped with Cs<sup>+</sup> and Rb<sup>+</sup>



## Ion-migration-retarding mechanism (experimental and theoretical)

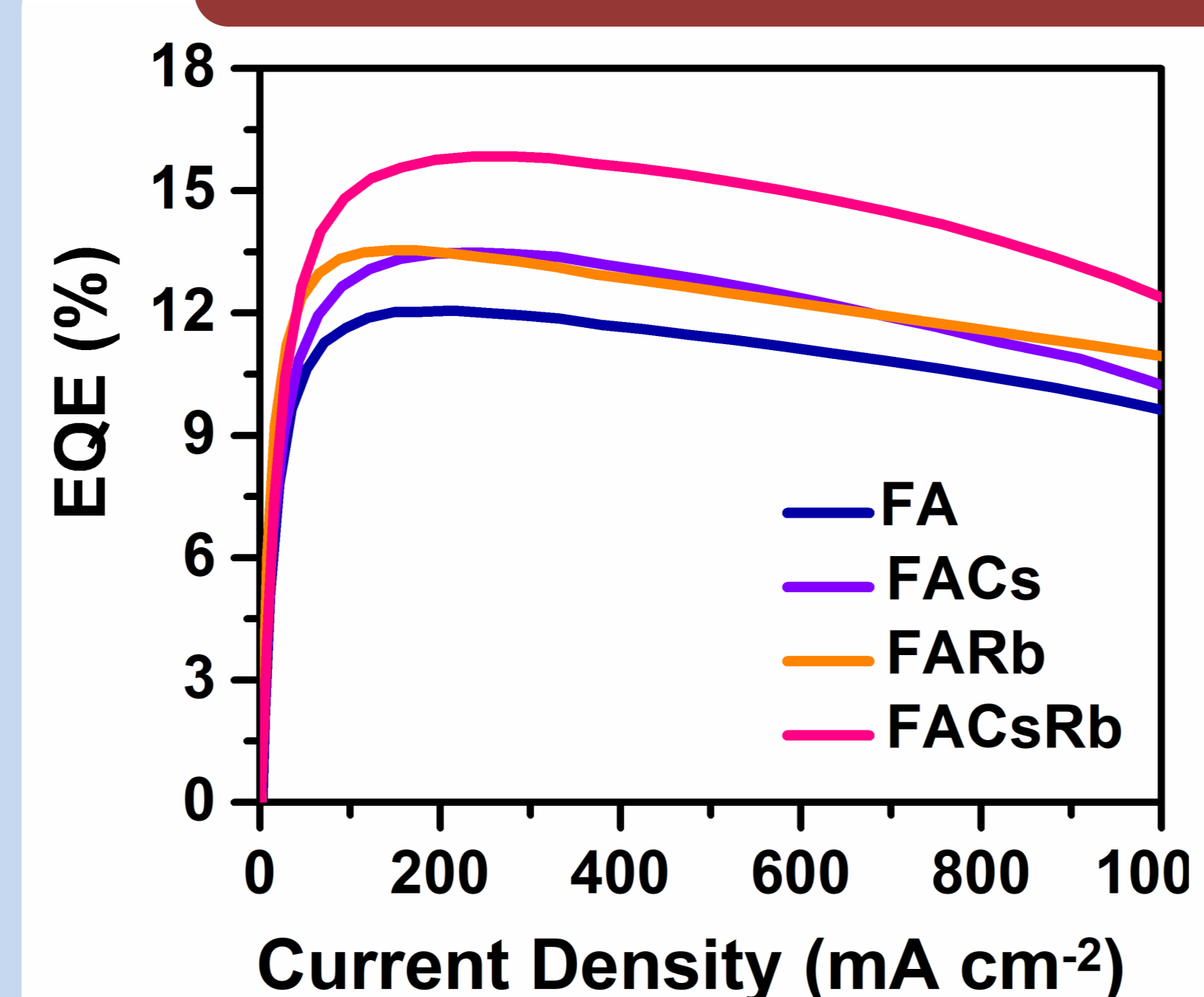


1. Cs<sup>+</sup> throughout bulk, Rb<sup>+</sup> on surface and GBs;
2. Both raising net atomic charge of I<sup>-</sup>, enhancing Coulomb interactions between cations and inorganic framework

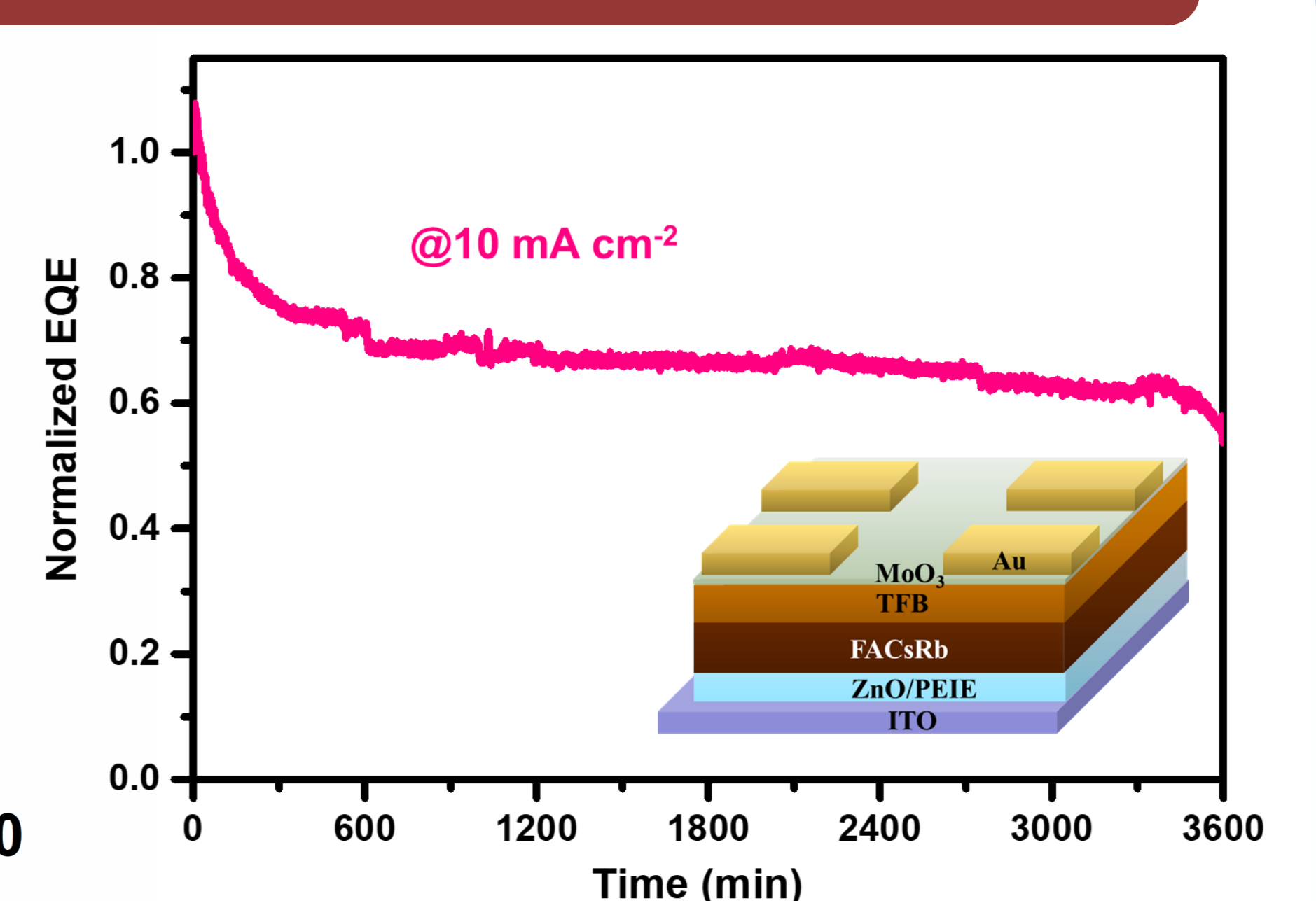
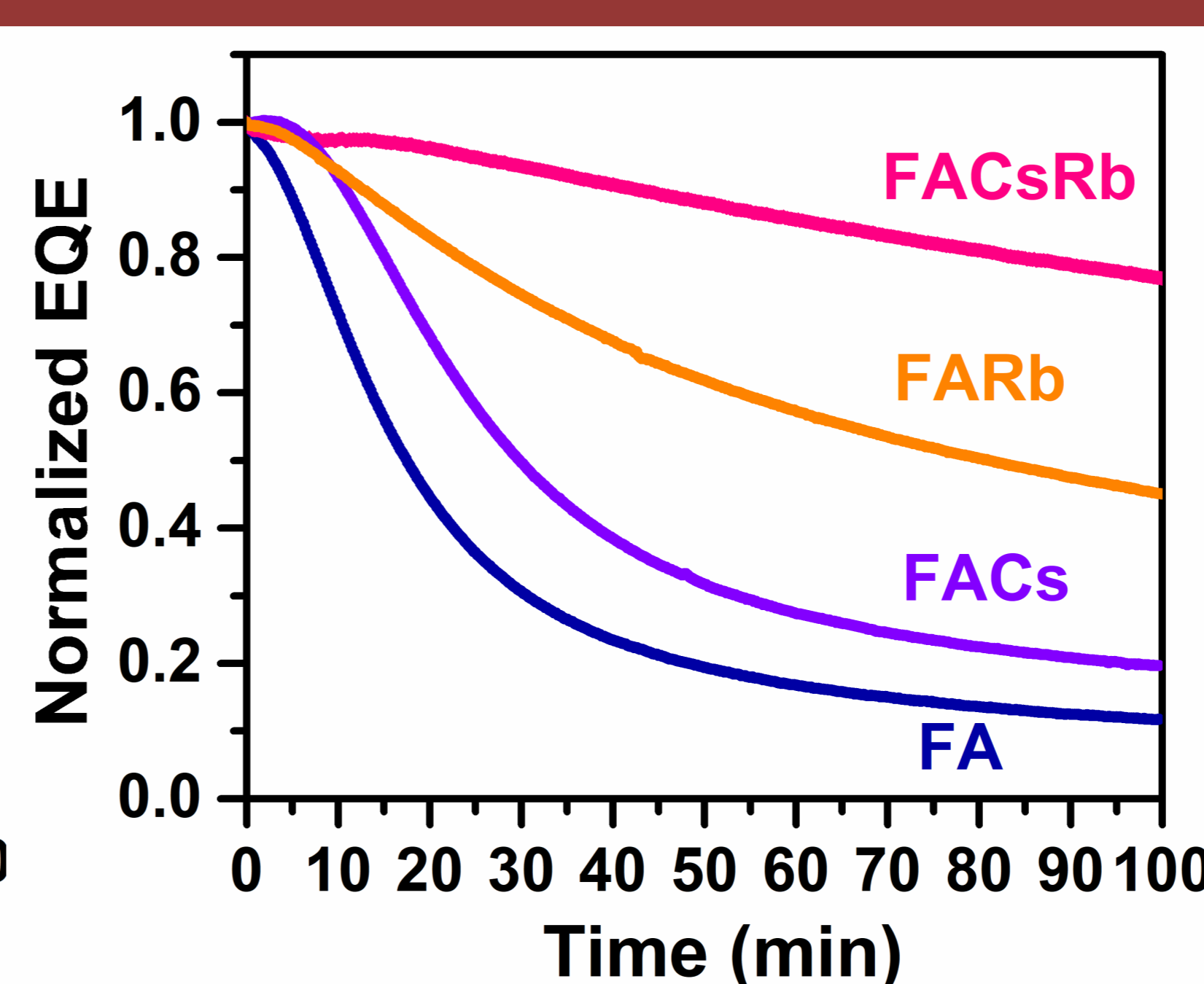


Improved film quality with reduced defects by Cs<sup>+</sup>-Rb<sup>+</sup> doping

## Optimized device performance and operational stability



Enhanced EQE up to 15.84% with comparable T<sub>50</sub> (over 3600 min) to NIR OLEDs



## Conclusion

We examined the degradation mechanism of FAPbI<sub>3</sub>-based PeLEDs during operation and explored ways of improving the PeLED stability via composition engineering. TOF-SIMS depth profiling shows severe accumulation of I<sup>-</sup> ions at the MoO<sub>3</sub>/Au interface, providing a direct evidence of ion migration during PeLED operation. By incorporation of Rb<sup>+</sup> and Cs<sup>+</sup> in FA perovskites, both film quality and device performance are significant improved. Rb<sup>+</sup> ions primarily locate at the grain boundaries and surfaces while Cs<sup>+</sup> mainly distribute in the bulk, both increase net atomic charges of I<sup>-</sup> anions and enhance the Coulomb interaction between cations and the inorganic framework, potentially retarding the formation of I<sup>-</sup> vacancy and blocking the I<sup>-</sup> migration both in the bulk and at GBs. We obtain a record EQE<sub>max</sub> of 15.84%, one of the highest among alkali-cation-incorporated FAPbI<sub>3</sub>-based PeLEDs. The device also shows superior durability with a T<sub>50</sub> over 3600 min, comparable to that of the NIR OLEDs. Our work provides a new approach to the fabrication of highly efficient and stable PeLEDs.

## Acknowledgement

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