

Ultrafast electroabsorption spectroscopy study of carrier dynamics within hybrid lead halide perovskites

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Since their first advent, hybrid lead-halide perovskite solar cells (PSCs) have kept their promises and proven to be a valid alternative to current solar cell technologies, with power conversion efficiencies rising above 20%. At this point, however, further improvements require a more fundamental understanding of some key features inherent to PSCs, such as their interfacial properties. Indeed, on top of travelling smoothly across the perovskite layer, the photocarriers must be injected efficiently into their respective extracting layers, for optimized devices.

Herein, we aim at unravelling the origin of performance inhomogeneity in PSCs by probing three essential properties : (i) the transmission of carriers across various heterojunctions, namely perovskite-*spiro*-MeOTAD and perovskite-SnO₂, (ii) the carrier recombination in the bulk material, and (iii) the carrier transport across the perovskite layer. In this respect, time-resolved electroabsorption spectroscopy (TREAS) appears as a method of choice. This technique relies on the ultrafast monitoring of the electroabsorption signal and allows to probe field-induced phenomena such as the dynamics of field screening and charge accumulation at interfaces. As a consequence, the three abovementioned properties can be successfully accessed when using this technique in combination with transient absorption spectroscopy (TAS).

In this study, two different perovskite materials (MAPbI₃ and (MA, FA)PbI_{3-x}Br_x) within four PSCs architecture exhibiting different interfacial structures have been studied by TREAS and TA spectroscopy techniques. We report a significantly more efficient electron injection at the SnO₂/(MA, FA)PbI_{3-x}Br_x interface, together with a decreased bulk recombination for the latter material, compared with the standard MAPbI₃. This accounts for the reported higher V_{OC} and altogether better performance of (MA, FA)PbI_{3-x}Br_x-based PSCs. At the material level, we assign this to the presence of nano-domains of various compositions favouring charge separation.