Charge Carrier and Exciton Dynamics in Mixed-Halide Perovskites

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Heterojunction solar cells based on hybrid organic-inorganic lead halide perovskites of various chemical compositions and nanostructures have been developed during the last decade. Devices based on thin-films of these materials currently achieve solar power conversion efficiencies exceeding 23 % and an open-circuit voltage of more than 1.2 V. Despite a number of intrinsic drawbacks, the exceptional photovoltaic performances of lead perovskites justify a detailed investigation of the mechanisms and dynamics of underlying phenomena.

The constituents of perovskite solar cells (PSCs) have significantly evolved since the first photovoltaic devices made out of the standard methylammonium lead triiodide (MAPI, $CH_3NH_3PbI_3$) to the latest developments relying on mixed cations, mixed anions perovskite systems and arrangements of 2D-3D layers. Indeed, it emerges from the last three years that mixed-composition perovskites and complex structures including a low-dimensional perovskite layer on top of a bulk three-dimensional perovskite film can perform significantly better than standard systems.

We use a combination of ultrafast spectroscopy techniques to scrutinize the carrier and exciton dynamics in standard and mixed-cations, mixed-halide lead perovskite thin films. The effects of photon excess energy and compositional variations on the mobility of hot carriers and its temporal evolution is probed by application of ultra-broadband THz spectroscopy. On the other hand, photoinduced transient Stark signals evidence the formation of charge transfer excitons (CTE) across the boundaries of nanodomains of various dimensionality and halide compositions.

These findings show that vectorial charge separation takes place in the film and at interfaces, which is at the origin of the improved efficiency of PSCs based on mixed-halide and structured materials, when compared to the standard MAPI.