

Hot Carriers in Metal Halide Perovskite Solar Cells

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Metal halide perovskites (MHPs) are emerging as an intriguing class of solution-based semiconductors with significant potential for optoelectronic devices, in particular photovoltaics; thus, a fuller understanding of the carrier dynamics and stability of these materials is critical to the development of solar cell physics. In this study we investigate the triple halide perovskite (FA,Cs)Pb(I,Br,Cl)₃, which is known to be amongst the most stable MHPs available. This stability allows a comprehensive study of the hot carrier (HC) dynamics to be assessed at high fluence and at various temperatures in a device architecture.

A systematic set of measurements including simultaneous photoluminescence (PL) and current-voltage (J - V) measurements and transient absorption (TA) spectroscopy were performed as a function of both power and temperature to probe the HCs and electron-phonon coupling in operational solar cells. These measurements support the presence of HCs in devices during operation, independent of any negative effects due to parasitic halide segregation or decomposition.[1] Indeed, the competition between HC dynamics and photo induced halide segregation are shown to be strongly dependent upon temperature. At 100 K, clear evidence of HCs is observed via increasing carrier temperatures in the high energy tail of the PL coupled with ballistic transport in J - V measurements. At room temperature, however, a blue shift in the PL and high energy broadening characteristic of HC generation at higher laser excitation are shown to compete with a gradual red shift in the PL peak and linewidth broadening as photoinduced halide segregation begins to occur at higher lattice temperatures.

The presence and properties of HCs is also presented in the ultrafast regime using TA spectroscopy. In this case, the dynamics of HCs are assessed in a device with upper and lower ITO electrodes to enable photoexcitation in the structure in the region of both the electron-transporting and hole transporting interfaces in the device. These devices were assessed at operational conditions with the HC thermalization probed at the maximum power point, short circuit current, and open circuit voltage conditions. Two distinct cooling (decay) times are observed when exciting at the two interfaces. The first decay which is a fast process, has a timescale of 5 ps. This is followed by a much slower decay that extends to hundreds of ps.[2] These data are discussed with respect to non-equilibrium carrier thermalization and lattice heating, coupled with non-equivalent carrier extraction relative photoexcited carrier absorption and density, as well as the role of the so-called phonon bottleneck in these systems.

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References

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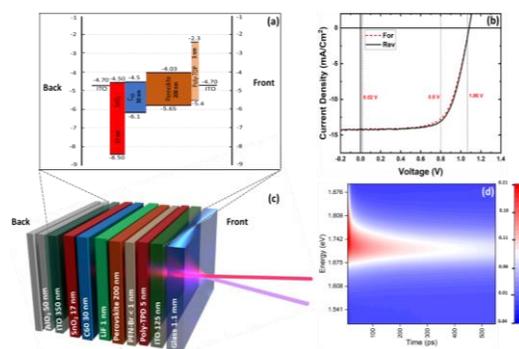


Fig.1. HCs are assessed in an operational MHP solar cell of structure shown in (a) and (b). The quality of this devices is reflected in the lack of hysteresis and retention of performance after each measurement (b). TA is assessed under fixed biases reflecting the maximum power, V_{OC} and J_{SC} . An example of which is shown in (d)