Deciphering Ultrafast Charge Carrier Dynamics in $MAPbI_3$/(PbS/CdS) Hybrid Thin Polycrystaline Film Using Transient Absorption and THz Spectroscopies

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Perovskite/(core shell) semiconductor quantum dots (QDs) have emerged as smart hybrid materials for light and energy conversions.[1-4] However, the real-time dynamics of photogenerated charge carriers is not yet understood. Here, interrogating a hybrid film of methylammonium lead triiodide ($MAPbI_3$) and PbS/CdS interface, we decipher the electron-hole events by using ultrafast transient absorption and terahertz spectroscopies.[5] We explored the effects of both pump wavelength and fluence on the mechanisms involving electron-hole relaxation, transport, trapping and recombination. We found that exciting the perovskite over its band gap, electron/hole transfers to/over the interface within 40 ps (Figure 1). We also observed deactivation processes having times of ~5 ps and 390 ps, assigned to excitonic states at the interface, and recombination over the band gap of perovskite, respectively. Interestingly, the perovskite-edge band gap excitation shows a significant influence of Wannier-Mott excitons dissociation and presence of local potential fluctuation on charge carrier transitions at the interface[6]. Our results decipher details of the working mechanism, and involved times of the related events, which play a key role in limiting the efficiency of these hybrid materials when used in photonics.

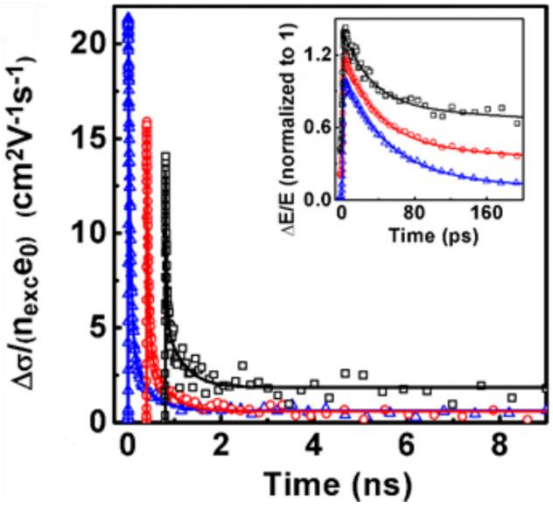

Figure 1: Photoinduced mobility decay of a $MAPbI_3$/QDs thin polycrystalline film upon excitation at 600 nm, using different fluences of the absorbed photon: 1.5×10^{12}, 8.2×10^{12} and 16×10^{12} ph/cm^2.

Funding: This work was supported by MINECO (MAT2014-57646-P, MAT2016-76892-C3-1-R), JCCM (PEII-2014-003-P) and Universitat Jaume I (UJI-B2016-05).

Acknowledgement: Mario Gutiérrez thanks the MINECO for the FPI fellowship.

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