

The origin of high photovoltaic efficiencies in large-grain organic-inorganic perovskites

J.-C. Blancon,^{a+} W. Nie,^a A. Neukirch,^a S. Tretiak,^a L. Cognet,^b A. Mohite,^a J. Crochet^a

^a *Los Alamos National Laboratory, Los Alamos, NM, 87544, USA.*

^b *Institut d'optique & CNRS, and Universite de Bordeaux, Talence, 33400, France.*

Organometallic perovskites have attracted considerable attention after promising developments in energy harvesting and other optoelectronic applications. Notably, the recent synthesis of high-crystallinity thin films with grain sizes of hundreds of microns has offered a new opportunity for the development of efficient solar cells with high reproducibility and photo-stability [1][2]. However, it is crucial to obtain a deeper understanding of the intrinsic photo-physics and transport properties of perovskites with relevant structural characteristics. Here, we will present an investigation of the dynamics of photogenerated charge carriers in large-area grain organometallic perovskite thin films via confocal time-resolved photoluminescence spectroscopy. Our findings show that the bimolecular recombination of free charges is the dominant decay mechanism at excitation densities relevant for photovoltaic applications [3]. Bimolecular coefficients were found to be on the order of 10^{-9} cm³/s, comparable to typical direct-gap semiconductors, yet significantly smaller than what is theoretically expected. We also will provide evidence that there is no degradation in carrier transport in these thin films due to electronic defects through modeling of the photoluminescence kinetics and open circuit voltage characteristics of a photovoltaic cell. We conclude that suppressed electron-hole recombination and transport that is not limited by defects provide a microscopic model for the superior performance of large-area grain hybrid perovskites for photovoltaic applications.

⁺ Author for correspondence: jblancon@lanl.gov

[1] W. Nie, et al., *Science* **347**, 522 (2015).

[2] W. Nie, et al., *Nat. Commun.* **7**, 11574 (2016).

[3] J.-C. Blancon, et al., *Adv. Func. Mater.* **26**, 4283 (2016).