Multiple exciton generation rate in $CsPbI_3$ quantum dots

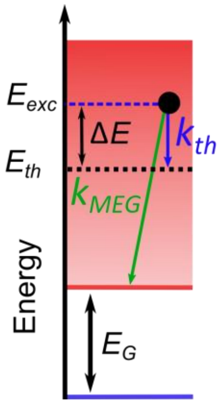
Osaka Univ., D. Timmerman, M. Ashida, and Y. Fujiwara

Osaka Dental Univ., E. Matsubara

Amsterdam Univ., L. Gomez, and T. Gregorkiewicz

E-mail: dolf.timmerman @mat.eng.osaka-u.ac.jp

The multiple exciton generation (MEG) process has been extensively investigated in a wide variety of quantum dots as it possesses the ability to positively disrupt solar cell conversion efficiencies. After absorption of high energy photons, hot excitons are formed in quantum dots, which will subsequently cool down to the band edge. The splitting of a hot exciton into two, or more, lower energy excitons, so-called MEG, is also possible. In general, the carrier generation quantum yield (QY) is determined by the competition of the exciton cooling processes and the MEG rate (Fig. 1). Although the determination of cooling rates is well-established, a direct observation of the MEG rate has so far been lacking, resulting from the short timescales and fast competing cooling processes. Perovskites have been found to have slower carrier cooling rates when compared to more conventional semiconductors, and also possess efficient MEG properties. Here, we present a study on the ultrafast carrier dynamics of $CsPbI_3$ quantum dots (Fig. 2), and determine both the exciton energy cooling and MEG rates. We find an exciton energy cooling time of 1 eV/ps and MEG rate of 2.5 s^{-1}, respectively. The MEG process is also observed directly in the induced absorption dynamics, as a delayed carrier concentration builds up following the excitation pulse (Fig. 3). These results underlie the high efficiency of MEG yields found for perovskite QDs, and present a new way of determining MEG characteristics.



$$QY = \frac{2k_{MEG} + k_{th}}{k_{MEG} + k_{th}}$$

Fig. 1 Exciton cooling model showing competition between MEG rate (k_{MEG}) and cooling (k_{th})

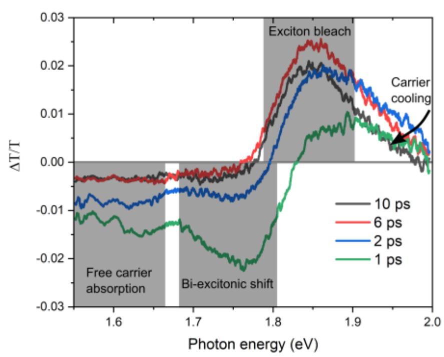


Fig. 2 Transient absorption for the first 10 ps after excitation of pump photon energy of 3.9 eV.

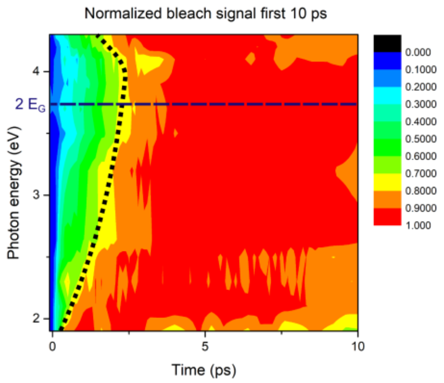


Fig. 3 Bleach signal for first 10 ps ater pulse for different pump photon energies