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Broadband ultrafast transient absorption of multiple exciton dynamics in lead sulfide nanocrystals FELICE GESUELE, CHEE WEI WONG, Columbia University, MATTHEW SFEIR, JAMES MISEWICH, Brookhaven National Laboratory, WEONKYU KOH, CHRISTOPHER MURRAY, University of Pennsylvania — Multiple exciton generation (MEG) is under intense investigation as potential third-generation solar photovoltaics with efficiencies beyond the Shockley-Queisser limit. We examine PbS nanocrystals, dispersed and vigorously stirred in TCE solution, by means of supercontinuum femtosecond transient absorption (TA). TA spectra show the presence of first and second order bleaches for the 1Sh-Se and 1Ph-Pe excitonic transition while photoinduced absorption for the 1Sh,e-Ph,e transitions. We found evidence of carrier multiplication (MEG for single absorbed photon) from the analysis of the first and second order bleaches, in the limit of low number of absorbed photons (Nabs$\sim$0.01), for energy three times and four times the Energy gap. The MEG efficiency, derived from the ratio between early-time to long-time TA signal, presents a strongly dispersive behavior with maximum red shifted respect the first absorption peak. Analysis of population dynamics shows that in presence of biexciton, the 1Sh-Se bleach peak is red-shifted indicating a positive binding energy. MEG efficiency estimation will be discussed with regards to spectral integration, correlated higher-order and first excitonic transitions, as well as the nanocrystal morphologies.

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