Ultrafast Relaxation Dynamics in Perovskite Nanostructures

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Hybrid perovskite materials are currently the subject of intense research regarding their use in solar cells since the reported power conversion efficiency has dramatically increased from 3.8% (2010) to 23.7% (2018). The performances of such devices are mainly limited by the energy losses of the photoactive material itself (ultrafast relaxation to the band-edge) and the difficulties to optimize the carrier extraction. A better understanding of the ultrafast relaxation dynamics of hybrid perovskite materials is thus of particular interest.

The first report of ultrafast relaxation in hybrid perovskite thin films evidenced the free carrier nature of the photo-excited species[1]. Moreover, a strong decrease of the cooling rate at high excitation densities was observed in methylammonium and formamidinium lead-iodide perovskites (MAPI and FAPI, respectively). This phenomenon was attributed to a "hot-phonon bottleneck" effect[2]. More recently, the development of colloidal synthesis of perovskite nanostructures has enabled the investigation of hot-carrier cooling in confined systems but these studies remain either limited to the weak confinement regime[3] or require a deeper characterization.[4]

In this work, we explore the effect of the quantum confinement and excitation density on the relaxation processes by studying three colloidal perovskites nanostructures synthesized in our laboratory: $FAPbI_3$ colloidal nanocrystals (weak 3D confinement) and strongly confined FAPI 2D nanoplatelets of one (n=1) and two (n=2) monolayers in thickness.

Using femtosecond transient absorption, we studied the hot-carrier relaxation to the band-edge after excitation at high energy and investigate how the cooling rate decrease for higher excitation density. From global analysis using Glotaran software[5], we differentiated the spectral signals resulting from the excitation above the bandgap (hot-carrier distribution in nanocrystals and pure Stark effect for nanoplatelets samples) versus the longer time signal corresponding to cooled down charge carriers (similar for excitation at the band-edge). We found stronger dependency of the pump fluency on the cooling rate for nanocrystals than for the 2D nanoplatelets and no clear effect of the presence of the organic cations.

References

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