Relaxation Dynamics of Hot Excitons and Charge Carriers in Colloidal Perovskite Nanostructures: Effect of Confinement

Carolina Villamil Franco[1], Benoît Mahler[2], Christian Cornaggia[1], Thomas Gustavsson[1] and Elsa Cassette[1]

[1]LIDYL, UMR 9222 CEA-CNRS, Université Paris-Saclay, CEA Saclay, 91191 Gif-sur-Yvette, France.

[2]ILM, UMR 5306 CNRS-Université Claude Bernard Lyon 1, 69622 Villeurbanne, France.

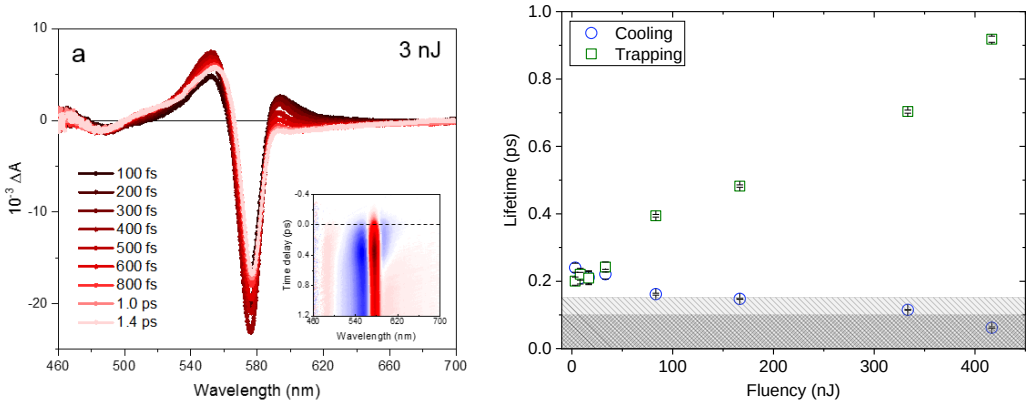

Figure. *Left: Time-dependent TA spectra of n=2 FAPI nanoplatelets (chirp corrected) with the full TA map in inset, for a pump excitation at 400 nm at low excitation density. Right: Excitation density dependent cooling and trapping lifetimes of n=2 FAPI nanoplatelets.*

The characteristic rate at which photoexcited hot charge carriers relax to the band edge has crucial implications on the performance of the optoelectronic devices. The dynamics and mechanisms of this charge carrier cooling in hybrid perovskite thin films have been the subject of intense research over the last few years. Here, we investigate the effect of confinement on the relaxation process of lead iodide-based perovskite nanostructures by comparing the cooling rate of weakly confined (thick nanoplates and large nanocrystals) and strongly confined 2D nanostructures (one and two monolayer thick nanoplatelets), using femtosecond transient absorption spectroscopy (TA).

Unlike the thick nanoplates which present bulk-like optical and relaxation properties, the strong quantum and dielectric confinement in the 2D nanoplatelets lead to discrete excitonic transitions near the bandedge, which prevent to use the classical model analysis for relaxation by extracting time-dependent carrier temperatures from a Boltzmann distribution. In addition, strong signals originating from Stark effects observed in the perovskite nanoplatelet TA spectra had to be isolated from the bleach ones. Using a global analysis method, we extracted the rates of charge carrier relaxation after pump excitation above the bandedge, at low and high excitation density. Trapping of one of the charge carrier to shallow states inside the bandgap was evidenced from the buildup of a low energy tail of the first excitonic transition bleach. Importantly, we found no delaying of the cooling process with the increase of the charge carrier density, per opposition with the strong “hot phonon bottleneck” observed in thin film (bulk) perovskite and 3D (bulk-like) nanocrystals (NCs). On the other hand, initially rapid (few 100s of fs) trapping slows down to picosecond by increasing the pump fluencies.