Element selective probing of the photo-induced charge carriers in inorganic lead perovskites

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Inorganic perovskite CsPbBr₃ nanocrystals are promising candidates as next-generation photonic sources for applications in displays or solid-state lighting. Despite the large research efforts in recent years, microscopic details of the charge carrier formation and relaxation are still missing. In this perspective, time-resolved X-ray absorption spectroscopy (TR-XAS) represents an ideal tool to investigate this multi-element material.

Previous time-resolved experiments performed at the Swiss Light Source (SLS) on the Br K-, the Pb L_3 - and the Cs L_2 -edges of CsPbBr₃ nanocrystals in toluene (optical pump 355 nm) have pointed out to localization of the holes at Br sites, which leads to formation of a small polaron in the valence band (VB) within 100 ps. The results also suggest that the electrons either remain delocalized or form large polarons in the conduction band (CB). [1,2]

Recently, these investigations were pursued at the Advanced Photon Source (APS, Chicago). The Br K-edge energy spectra at different time delays have been recorded to probe the local structural modification in the EXAFS region and kinetic traces at the pre-edge, edge and post-edge were collected to determine the relationship between the electronic dynamic at Br sites and the structural changes of the local environment. Furthermore, high-energy resolution fluorescence detection (HERFD) was implemented at the Pb L₃-edge XANES, yielding finer details of the broad featureless absorption. The study allows us to obtain further insights about the charge carrier relaxation process and relates the spectral features at the Pb centers with the nuclear and electronic changes of the Br sites.

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