Novel semiconductor nanocrystals: ultrafast spectroscopy and energy conversion

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Lead halide perovskites (LHPs) have developed to a highly promising material system for photovoltaic applications. Nanocrystals composed of LHPs allow for highly efficient light emitters tunable over the entire visible spectrum. The incorporation of the heavy metal lead, however, makes the material toxic and unstable when exposed to heat or moisture. Bismuth is a promising alternative, being stable, less harmful and as an ion isoelectric with lead. Double perovskites, bismuth vanadate and bismuth oxyhalides have recently attracted substantial interest for optically driven energy conversion applications such as photocatalytic water splitting. However, in many cases the underlying microscopic processes leading to favorable or less favorable performance are not understood in a satisfactory way.

We have performed a series of time-resolved optical spectroscopy studies in order to shine light onto the carrier dynamics occurring in colloidal nanocrystals made out of the above mentioned material systems. In all cases an understanding can only be achieved by knowing the electronic energy landscape, i.e. the band-structure of the studied semiconductor materials altered by quantization and Coulomb effects. For LHP nanoplatelets and LHP quantum dots I will discuss relaxation processes of charge carriers and spins. The analysis of coherent phonons and polarons in bismuth oxyhalides leads to an understanding of their size dependent performance in photocatalytic experiments. Finally, an indirect band-gap together with high effective masses and trap states for holes lead to an explanation of the unique carrier relaxation scenario in double perovskites.