Inorganic and organic semiconductor nanocrystals for photocatalytic water splitting

Jacek Stolarczyk

Photocatalytic generation of fuel holds promise to simultaneously address two main challenges in energy conversion: harvesting and storage. To this end, the energy of incident photons is utilized to drive energetically up-hill reactions of water splitting or CO_2 reduction. Semiconductor nanocrystals, both inorganic and organic, have the potential to absorb light, separate the photoexcited charge carriers and induce the desired surface reactions.[1,2] In the talk, I discuss the efforts to understand and control the dynamics of the charge carriers and thereby to increase the photocatalytic efficiency.

Ultrafast spectroscopic experiments show that by careful management of charge carrier transfer rates and recombination rate, the quantum efficiency of H_2 generation on metal-decorated CdS nanorods can exceed 90% under blue light illumination.[3] Combining the nanorods with oxidation catalysts, full water splitting is demonstrated on a single particle under visible light illumination.[4]

Carbon dots are versatile and very promising materials for photocatalysis and light emission but the inherent complexity of their structure makes it difficult to provide clear design guidelines. It is shown that the model of polycyclic aromatic hydrocarbons embedded in an amorphous matrix, supplemented by individual molecular fluorophores, accounts for the unusual optical properties of the dots. Their function can be switched between photocatalytic and emissive properties through simple preparative means.[5] Furthermore, the photobasic effect of nitrogen-containing aromatic molecules is demonstrated to enhance the hydrogen production by pulling the proton to the reaction site on a picosecond timescale, precisely when needed.[6]

In the final part, I will discuss highly anisotropic inorganic nanocrystals. Ultrathin organometal lead halide perovskite nanoplatelets are reported with optical properties finely controlled by quantum confinement effects and the exciton binding energy. It is demonstrated how an internal anisotropy promotes charge separation. In bismuth oxyiodide nanoplatelets made of alternating charged layers, the ultrafast separation into different layers launches coherent phonons in its wake, providing a perfect start for efficient photocatalysis.[7]

References

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