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GROWTH, STRUCTURE AND FUNCTIONALITY OF SEMICONDUCTOR NANOCRYSTALS THROUGH Z-CONTRAST MICROSCOPY AND THEORY

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It is well known that quantum confinement effects in semiconductor nanocrystals are the origin of the size-dependent electronic and optical properties and their many potential applications in optics, electronics and catalysis. However, to understand nanocrystal functionality at a fundamental level requires more detailed knowledge of surface termination, stoichiometry and the presence of special sites such as vacancies that might act as traps for carriers or active sites for catalysis. The combination of atomic-resolution Z-contrast scanning transmission electron microscopy (STEM), electron energy loss spectroscopy (EELS) and first-principles theory is very effective in unraveling the origin of functionality in nanostructures. Z-contrast imaging of nanocrystals allow reconstruction of the three-dimensional shape. In the case of chemically-synthesized CdSe nanocrystals, the shape is asymmetric reflecting the polarity of the crystal. Images of core shell nanocrystals show directly the extent of the core. Such information is an excellent starting point for first-principles theoretical studies. As an example, we show that photocatalytic fixation of CO_2 by CdSe nanocrystals does not occur at a surface site at all, but by the *injection* of highly reactive CO_2^- radicals into solution where they can react and “fix” other CO_2 molecules. Determining nanocrystal functionality will be greatly facilitated by the reduced probe sizes after correction of objective lens spherical aberration, when we anticipate single atom sensitivity for imaging and perhaps also for the detection of impurities or dopants by EELS.