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Microsecond-resolved insights by SAXS and WAXS into the early stages of CdS quantum dot formation

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Semiconducting nanoparticles (quantum dots) show a wide range of potential applications due to their unique size-dependent physical and chemical properties. A major issue today concerns the make of such particles with a sufficient control of the particle size, shape and polydispersity, which calls for a good understanding of the formation mechanisms involved. We have developed a free liquid jet setup which allows to access a so far unexplored time regime from 20 μ s up to 10 ms. The key advantages compared to capillary based outfits are: 1) access of very early stages (1000 times faster than in stopped-flow experiments), 2) high time resolution (down to 10 μ s), 3) no radiation damage in the sample, and 4) high quality data evaluation because of missing container scattering. For longer timescales we have pioneered a free drop setup, which again provides for container-free measurements for reaction times beyond 100 ms. By SAXS experiments the sizes and morphology of the early particle states are accessible while simultaneously acquired WAXS patterns give insights into the evolution of the crystalline structure.

Both SAXS and WAXS studies show, that the CdS quantum dot formation from aqueous solutions is along a non-classical two-step nucleation pathway starting with the formation of primary clusters driven by the fast ion diffusion. Further growth is by cluster attachment where the diffusion of the primary clusters appears as the growth-limiting factor. Temperature dependent data yield a diffusion with an activation energy of $E_g=0.6$ eV.

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