

Synthesis of low-strain doped colloidal quantum dots

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A decade has passed since the introduction of the organometallic synthesis of colloidal semiconductor nanoparticles, also known as colloidal quantum dots (cQDs), by Murray *et al* [1], but it is still challenging to create high quality cQDs with a broad variety of stable photoluminescence (PL) properties [2]. To improve stability, our approach combines core/multishell cQDs separating the core from the environment with doping agents in the core such as divalent metal cations. Among the wide variety of synthetic protocols for II-VI cQDs, we opted for a non-coordinating solvent with fatty acid complexes as metal precursors.

The main idea behind core/multishell cQDs compared to the single shell version is to judiciously choose core and shells that will provide the smallest lattice mismatch between them [2][3]. A typical core of CdSe can be successfully isolated from the environment by consecutive shells of CdS, Cd_{0.5}Zn_{0.5}S and finally ZnS. The PL of these well-passivated low-strain cQDs is very resistant to photobleaching and ligand exchange. Moreover, the so-called cQD blinking is partially suppressed [4][5]. These characteristics make low-strain cQDs good candidates as labels for single biomolecule imaging, but the remaining challenge is to reduce their size to avoid perturbing molecular dynamics.

Here we propose to reduce the strain in the core/multishell cQDs even further by using Cd_xMg_{1-x}Se alloys which are better lattice-matched to CdSe while keeping an energy gap as wide as ZnS. We first investigated different Cd:Mg ratios in the shell alloys as well as different synthetic methods. For example, a procedure for CdSe/Cd_{0.8}Mg_{0.2}Se core/shell cQD synthesis starts by mixing and degassing octadecene (ODE) and oleylamine with already synthesised and washed core CdSe cQDs. The mixture is then heated to 250°C under argon flow and small volume of 0.1 M of Se in ODE is added to the reaction flask. After a sufficient reaction time

of about 10 minutes, the same small volume of a solution of both 0.02 M magnesium oleate and 0.08 M cadmium oleate in ODE is added. Each successive injection of chalcogen precursor followed by the metal precursor injection form a monolayer and this is repeated to make about nine monolayers of the alloy. This method creates very crystalline and monodisperse cQDs. In figure 1, the core and shell of the cQDs have nearly identical lattice parameters and the magnified particle clearly shows a regular lattice with no apparent dislocation.

We have also begun investigating insertion of divalent metal cations like Mn²⁺ in the CdSe core before covering them with Cd_xMg_{1-x}Se.[7][8]

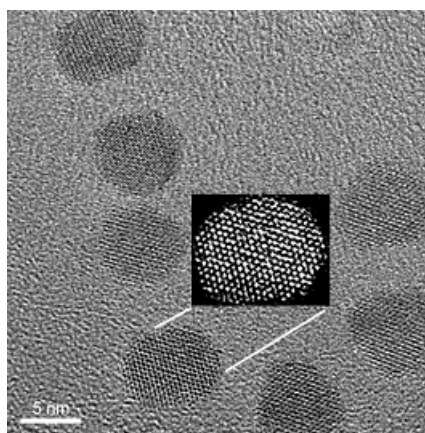


Figure 1: Transmission electron micrography of CdSe/MgSe cQDs. In the inset, magnification with enhanced contrast of one cQD

References

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