Nanoscale area selective ZnO growth between a monolayer of nanocrystals

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Colloidal semiconductor nanocrystals or quantum dots (QDs) combine a broad absorption spectrum with a narrow, highly efficient and tunable emission. Therefore they are actively investigated for applications in opto-electronic devices such as light emitting diodes, amplifiers or lasers and photovoltaic cells. For many applications, QDs need to be embedded in a solid matrix, either to reduce degradation due to exposure to moisture and oxygen or to allow efficient injection or extraction of electron-hole pairs.

Here, the encapsulation of a monolayer of CdSe/CdS/ZnS core/shell QDs in a ZnO matrix by ALD is studied. A monolayer of core/shell QDs with an overall diameter of 10 nm that are capped with oleate ligands was formed on a silicon substrate by Langmuir-Blodgett deposition. This ensures the deposition of a single monolayer of QDs, ordered in a hexagonal pattern. The encapsulation of the QDs by ALD of ZnO was monitored in situ through synchrotron based X-ray fluorescence (XRF) and grazing incidence small angle scattering (GISAXS) measurements. The series of peaks in the GISAXS data (Fig. 1a) provide a direct confirmation of the local, hexagonal ordering within the monolayer of QDs. As the position of these peaks does not shift during ALD, we conclude that the encapsulation process (involving substrate heating and ALD) maintains the original order, i.e. QDs do not coalesce and/or melt during the ALD treatment. The XRF measurements allow us to monitor the number of deposited Zn atoms. The integrated intensity of the Zn Kα line is depicted in Fig. 1b. During the first 80 ALD cycles, a lower amount of ZnO is deposited on the QD sample as compared to a planar reference substrate. This points towards area selective growth, where ZnO is only deposited in the interstices in between the QDs, while no ZnO grows on the oleate capped QDs. After 80 cycles, which would corresponds to a layer thickness of about 15 nm, the growth rate corresponds to that on a planar reference substrate, as can be expected once the QDs have been overgrown by ZnO.

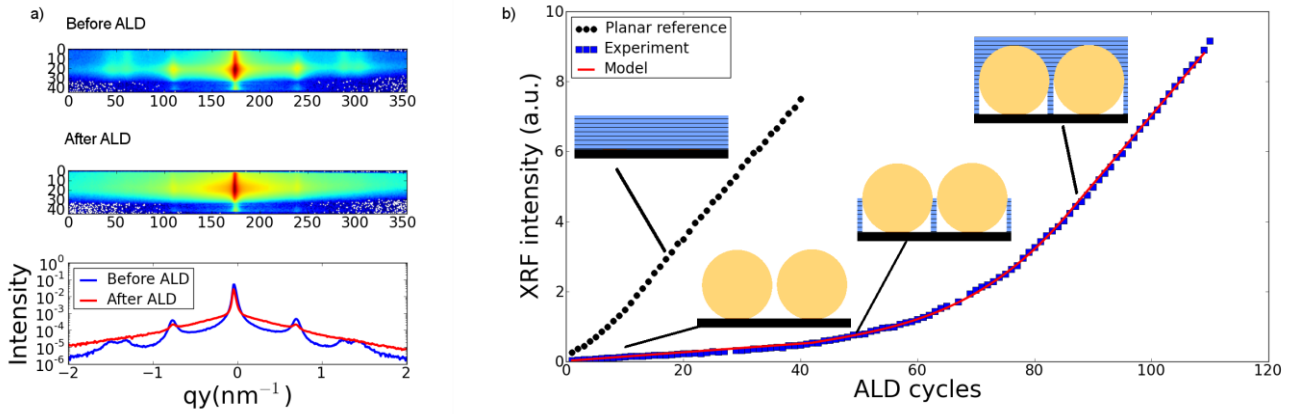

Fig.1 a) GISAXS pattern before and after 100 ALD cycles of ZnO deposition, showing the hexagonal order of the QDs. The bottom graph shows a cross-section at the Yoneda angle. b) XRF intensity as monitored in situ during ALD growth, showing a reduced amount of deposited material on the monolayer of QDs during the first cycles in comparison to the growth on a planar surface, suggesting that growth is only occurring in the interstices in between the QDs. The red curve shows a fit of the in situ XRF data based on these assumptions.