In situ synchrotron based XRF and GISAXS study of ALD encapsulation of supported nanocrystals

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Colloidal semiconductor nanocrystals or quantum dots (QDs) 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. ALD is a prime candidate to perform this task.

Here, the encapsulation of CdSe/CdS/ZnS core/shell QDs in a ZnO matrix is studied by in situ synchrotron based x-ray fluorescence (XRF) and grazing incidence small angle x-ray scattering (GISAXS). GISAXS clearly shows the order present in the monolayer of Langmuir-Blodgett deposited QDs, both before and during the encapsulation process. The XRF data is used to monitor the amount of deposited material. No ZnO growth occurs directly on the QDs during the first 50 ALD cycles. Previous studies, however, have shown that it is possible to use thermal ALD of Al₂O₃ to encapsulate similar QDs.[1] Therefore, a pretreatment of the QDs with TMA was used to facilitate the ZnO growth. During the exposure to TMA, a reduction of the Zn content is observed, indicating the etching of the ZnS shell of the QDs by the TMA. This pretreatment enables the subsequent growth of ZnO. However, a clear delay in nucleation is still present compared to the growth on SiO₂.



Figure 1 Gisaxs pattern before and after 48 ALD cycles of ZnO growth and TMA treatment, showing the conservation of order during encapsulation.



Figure 2 The integrated XRF intensity of the Zn K_{α} line indicating the amount of deposited ZnO during the encapsulation process. The growth on SiO₂ is added as a reference. The inset shows the loss of Zn form the ZnS shell of the QDs during TMA exposure of the QDs.

[1] K. Lambert, J. Dendooven, C. Detavernier, Z. Hens, Chem. Mater. 23 (2011). 126-128