In situ GISAXS during atomic layer deposition of oxides on ordered quantum dots

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Atomic layer deposition (ALD) is a self-limited growth method that is characterized by alternating exposure of the growing film to chemical precursors, resulting in the sequential deposition of (sub)monolayers [1,2]. The key advantages of ALD are the atomic-level thickness control and the excellent conformality, even on complex 3D nanostructures [3]. Here the coating and encapsulation of CdSe/CdS/ZnS core/shell quantum dots (QDs) in a ZnO, TiO2, Al2O3 and HfO2 matrix by ALD is studied. 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.

In this work, all depositions and measurements were performed in the UHV film growth facility, adapted for ALD, installed at beamline X21 of the National Synchrotron Light Source at Brookhaven National Laboratory. A monolayer or multilayer 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 layer of QDs ordered in a hexagonal pattern. The encapsulation of the QDs by ALD 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. 1) 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. XRF measurements allow us to monitor the number of deposited metal atoms in the oxide matrix. The combination of both techniques allows the study of the growth mechanism involved in the deposition of the oxide layers. Various oxides show a very different behaviour, which could be linked to differences in chemical reactivity between the ALD precursor gasses and the ligands or supporting substrate.

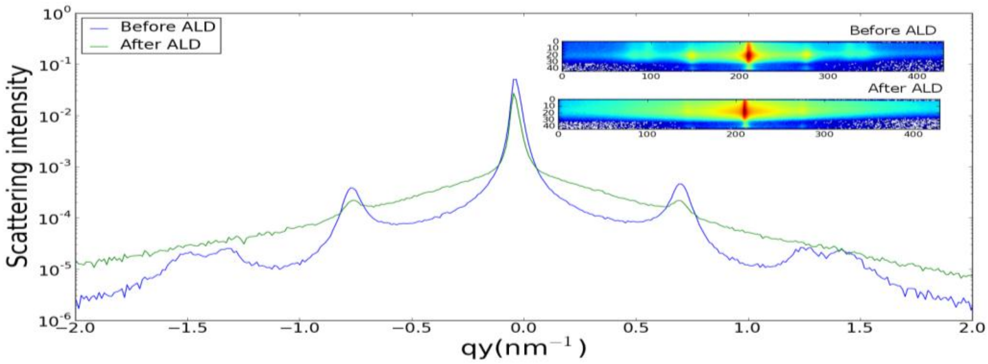

Figure 1: GISAXS pattern before and after 100 ALD cycles of ZnO deposition. The scattering intensity is shown at the Yoneda angle. The inset shows the 2D Gisaxs patterns. These show the hexagonal order of the QDs and the conservation of inter dot distances after deposition.

References

[1] S. M. George, Chem. Rev. **110**, 111 (2010)

[2] R.L. Puurunen, J. Appl. Phys. **97**, 121301 (2005)

[3] J. Dendooven, S.P. Sree, K.D. Keyser, D. Deduytsche, J.A. Martens, K.F. Ludwig, and C. Detavernier, J. Phys. Chem. C **115**, 6605–6610 (2011).