

## Investigation of GaN/AlN Quantum Dot Formation using Nanoscale Cathodoluminescence Microscopy

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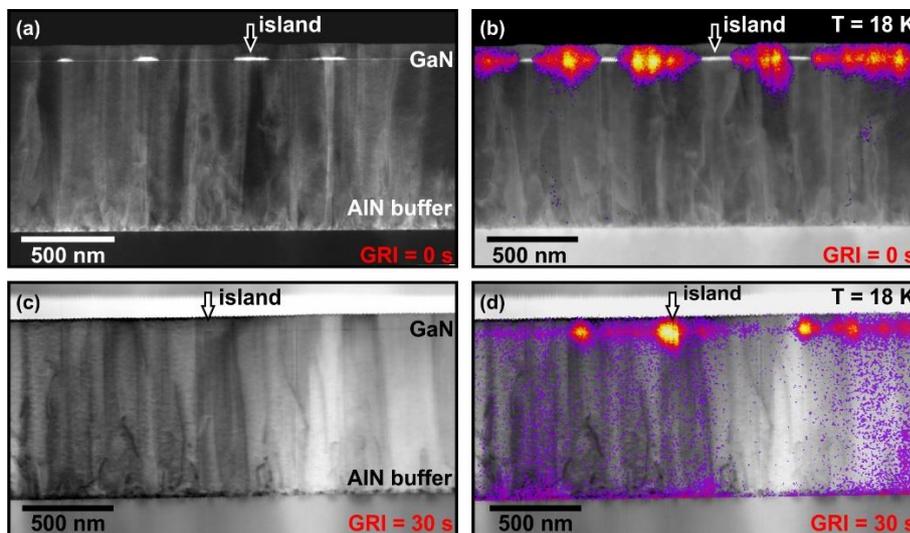
The generation of non-classical states of light is of fundamental scientific interest in quantum optics and an important technological challenge in quantum information processing. Semiconductor quantum dots (QDs) are prominent candidates for the realization of miniaturized single-photon emitting devices because of their atom-like energy structure and capability for electrically triggered operation. In particular, III-nitride QDs are promising room-temperature quantum emitters, for which large exciton binding energies and strong confinement are required.

We systematically studied desorption induced GaN/AlN quantum dot formation using cathodoluminescence spectroscopy (CL) directly performed in a scanning transmission electron microscope (STEM). The GaN films were grown by metal organic vapor phase epitaxy on top of an AlN/sapphire-template. After the deposition of few monolayers GaN at 960 °C a growth interruption (GRI) without ammonia supply was applied to allow for quantum dot formation. A sample series with GRIs from 0 s to 60 s was prepared to analyze the evolution systematically. Each structure was capped with AlN grown at 1195 °C.

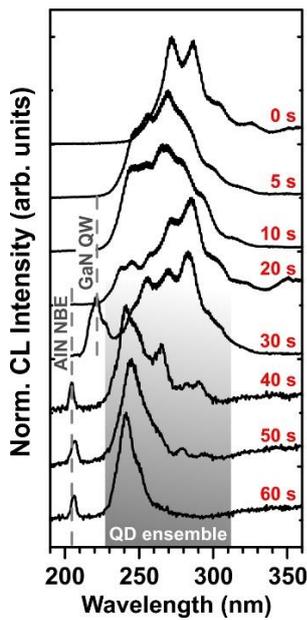
Without GRI cross-sectional STEM images of the reference sample reveal a continuous GaN quantum well (QW) with additional GaN islands of 20 nm height and ~100 nm lateral diameter nucleated at dislocation bundles. Spatially averaged spectra exhibit a broad emission band between 250 nm and 360 nm corresponding to the QW. In contrast, the GaN islands exhibit drastically reduced CL intensity.

Applying a growth interruption, desorption of GaN is observed resulting in smaller, fragmented islands in close vicinity to dislocations indicating desorption as the major influence. Now these islands exhibit ultra-narrow emission lines down to 440 eV in the range of 240 nm to 330 nm for 30 s GRI. Single photon emission is verified by a clear anti-bunching observed in Hanbury-Brown-Twiss photoluminescence experiments proving QD like emission.

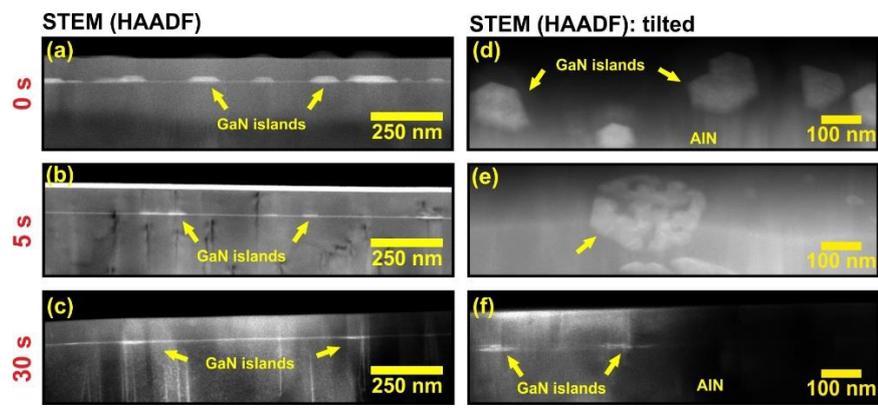
With increasing GRI, the QD as well as QW luminescence shifts to shorter wavelengths ending with 242 nm QD peak wavelength and decrease of the QW emission.



**Figure 1:** (a) Cross-sectional high angle annular dark field (HAADF) image of the sample with no GRI shows a continuous GaN film with GaN islands (marked with arrow) nucleated at dislocation bundles with (b) corresponding panchromatic CL image (color coded) superimposed with the STEM image shows CL intensity mainly from continuous GaN film and drastically reduced intensity at islands. (c) The bright field image of the sample with a GRI of 30 s exhibits small GaN islands as well as thin, continuous GaN quantum well is seen. (d) The recorded panchromatic CL image (color coded overlaid with the STEM image) displays strongly isolated and spot like emission dominantly from islands.



**Figure 2:** Spatially averaged CL spectra of the sample set with GRI from 0 s to 60 s taken at 6 K



**Figure 3:** (a-c) cross-sectional and (d-f) tilted HAADF images of the sample with (a, d) no, (b, e) 5 s and (c, f) 30 s growth interruption demonstrating the morphological change from thick, compact islands to smaller, fragmented islands ending in isolated GaN QDs for GRI  $\geq$  30 s