Cathodoluminescence spectroscopy of zinc-blende GaN quantum dots

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We analyzed the cathodoluminescence of zinc-blende GaN/AlN quantum dots grown by two different methods. Method A being droplet epitaxy, a vapor liquid solid process and method B being Stranski-Krastanov growth. At an equal quantum dot density of 10^{11} cm^{-2}, droplet epitaxy grown quantum dots have shown a luminescence intensity which was more than one order of magnitude higher than the one of Stranski-Krastanov quantum dots. Furthermore, we revealed a dependency between the quantum dot emission energy and the amount of deposited Ga.

1 Introduction Wide-bandgap quantum dots (QDs) based on group-III nitrides are of great interest for their possible applications in optical communication and quantum optical devices. Single photon emitters (SPE) based on wurtzite GaN single quantum dots have shown photon antibunching and triggered single-photon generation [1]. Built-in electric fields in the hexagonal phase of GaN induce a Quantum-Confined Stark Effect. This results in a low recombination probability of electrons and holes, which is undesirable for optical devices. Due to the high crystal symmetry, the metastable cubic phase of GaN (c-GaN) and AlN (c-AlN) has no polarization field in (001) growth direction [2]. Consequently the radiative recombination time of c-GaN QDs was measured to be two orders of magnitude lower than the recombination time of hexagonal GaN QDs [3].

We report on cathodoluminescence (CL) spectroscopy of molecular beam epitaxy (MBE) grown c-GaN QDs. Our QDs are embedded in c-AlN confinement layers on 10 µm 3C-SiC on top of Si (001) substrates. Two methods have been used to fabricate c-GaN QDs. Method A, droplet epitaxy (DE), is a vapor liquid solid process. Ga droplets are deposited on the AlN surface and subsequently nitridated [4]. Method B is based on the Stranski-Krastanov (SK) growth process. In this growth process the QDs are formed by relaxation of a pseudomorphically strained GaN layer [5, 6]. The emission energy of the quantum dots is tuneable by alternation of the quantum dot height.

Cathodoluminescence spectroscopy provides high sensitive analysis over a wide spectral range with high spatial resolution [7]. The principal advantage of CL is determination of the spatial resolution by the excess carrier generation volume. It is thus not limited by diffraction in the collection and excitation optics inherent in other spectroscopy techniques. Hence, CL is the method of choice to excite the c-AlN confinement (direct bandgap 5.93 eV, indirect bandgap 5.3 eV) [8] and investigate the optical properties of a single layer of c-GaN QDs. Furthermore, c-GaN QDs without c-AlN cap layer were studied by atomic force microscopy (AFM) to obtain data about the QD height, density, and size distribution.

2 Experimental procedure The c-GaN QDs and c-AlN confinement layer were grown in a Riber 32 MBE system including standard Ga and Al effusion cells and an Oxford Instruments N plasma source. The growth process was monitored *in-situ* by reflection high energy electron diffraction (RHEED). CL measurements were performed in a ZEISS DSM 950 scanning electron microscope equipped with an Oxford Instruments CL302 system and a CF301 helium cooled cryostat. Figure 1 shows a schematic drawing of this cathodoluminescence spectroscopy setup. The sample stage was docked to the cryogenic ring to investigate CL at low temperatures. To measure the sample temperature an AlGaAs temperature diode is located inside the sample holder. The sample emission was collected by
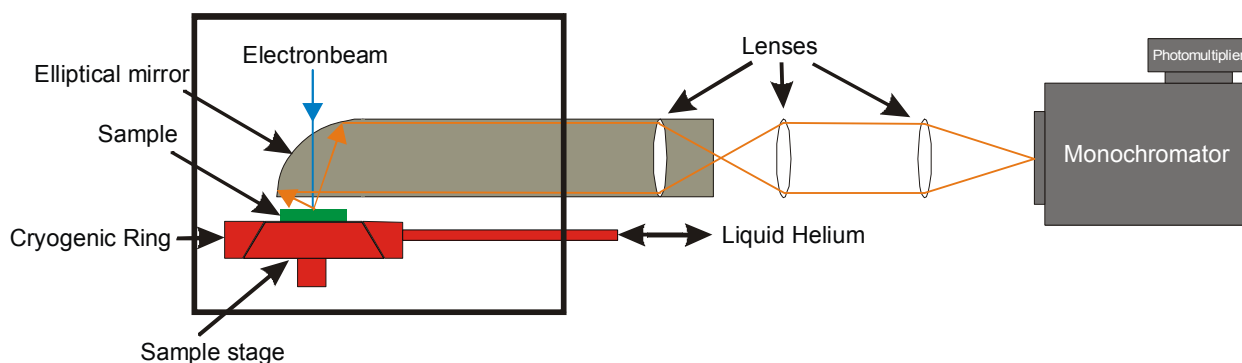
Figure 1 The schematic of the cathodoluminescence spectroscopy setup. Our CL system is based on an Oxford Instruments CL302 light collector and CF helium cooling cryostat is installed in a Zeiss DSM 950 scanning electron microscope (SEM). The collected light is guided by free optics to a monochromator and detected by a photomultiplier.

an elliptical half space collimator mirror and guided by free space optics. The spectrum separation was realized by a 0.15 m Princeton Instruments grating monochromator with variable entry and exit slits to adjust the ratio of spectral resolution and intensity. A Peltier cooled 1P28 photomultiplier operating in photon counting mode and a Hamamatsu photon counter C3866 was used for detection [9]. To obtain surface data of the uncapped QDs samples a Nanosurf Mobile S AFM system was utilized in contact mode.

3 Results and discussion

3.1 Growth Cubic GaN QDs are fabricated by droplet epitaxy and the Stranski-Krastanov growth process. The c-GaN QDs of both types are embedded in 30 nm c-AlN (001) confinement layers grown at 730 °C substrate temperature on 3C-SiC (001) by plasma assisted molecular beam epitaxy (PAMBE) [6]. In Fig. 2(a) the RHEED pat-

tern of the AlN buffer layer in [110] azimuth shows long thin streaks, that indicate a smooth two-dimensional surface [10]. QD fabrication by droplet epitaxy starts with the creation of Ga droplets at a substrate temperature of 350 °C. A defined amount of Ga, equivalent to 3-7 GaN monolayers (ML), is deposited on the c-AlN surface. The equivalent GaN ML are a criterion for the incorporated amount of Ga [4]. The strong cohesion force between Ga atoms induces the accumulation of Ga in droplets. To control the size and density of the droplets the substrate temperature and the amount of deposited Ga can be varied. Ga droplets are nitridated for 3 to 10 minutes while gradually ramping up the substrate temperature from 350 °C to 730 °C. Figure 2(b) shows the RHEED pattern after the nitridation of the Ga droplets. Electron transmission components through three-dimensional islands on the surface result in spotty reflections, which indicate QDs on the c-AlN layer.
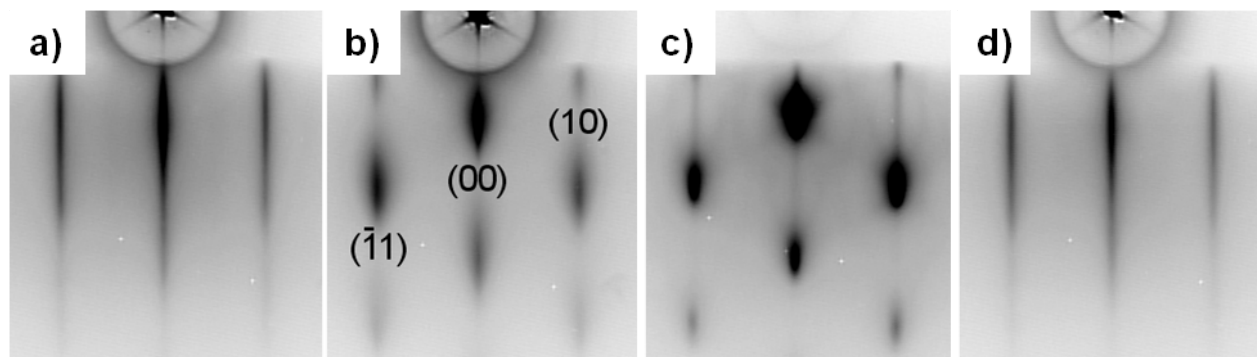

Figure 2 RHEED patterns: (a) Reflections of the c-AlN buffer layer, long thin streaks indicate a smooth 2D surface, (b) reflections of c-GaN DE QDs, the spotty reflections indicate 3D islands of cubic crystal structure, (c) reflections of c-GaN SK QDs and (d) reflections of the c-AlN cap layer after 30 ML QD overgrowth [6].
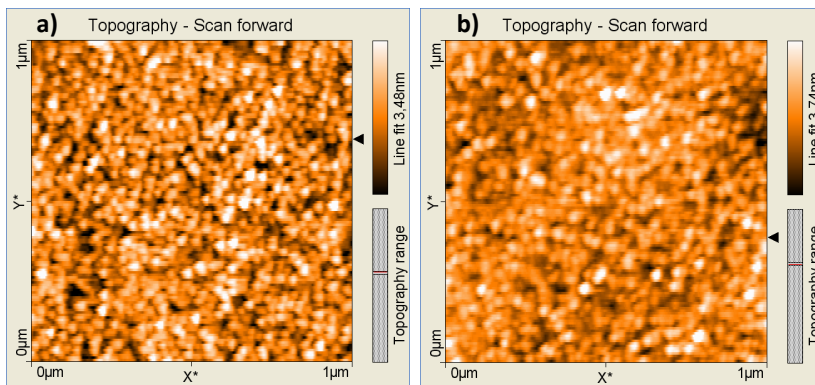
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Figure 3 AFM images: (a) 1x1 μm² area of DE c-GaN QDs on c-AlN. The average width is 20 nm, the height 3 nm and the density 1x10¹¹ cm⁻². (b) 1x1 μm² area of SK c-GaN QDs on c-AlN. The average width is 20 nm, the height 3 nm and the density 1x10¹¹ cm⁻².

The SK QDs are created by strain induced islanding in lattice-mismatched heteroepitaxy. The lattice mismatch between c-AlN and the c-GaN of 3.2 % [11, 12] results in formation of SK QDs after reaching a critical thickness of 2 ML GaN on c-AlN. In Fig. 2(c) the RHEED pattern after the formation of SK QDs is shown. Spotty reflections again indicate QDs on the c-AlN surface. The density and size of the QDs can be controlled by the amount of deposited GaN on the AlN buffer.

Finally a 30 nm c-AlN cap layer is grown to cover the c-GaN QDs. The RHEED pattern in Fig. 2(d) shows long thin streaks without spotty reflections. Therefore a complete overgrowth of the QDs and a smooth surface can be concluded. All reflections in the RHEED pattern of Fig. 2 indicate zinc-blende crystal structure.

3.2 Atomic force microscopy To investigate height and density of the QDs, uncapped c-GaN QDs samples were analyzed by AFM. The AFM image in Fig. 3(a) shows a 1x1 μm² area covered with DE QDs. The average width of the QDs is 20 nm and the height 3 nm. Figure 3 (b) shows an AFM image of 1x1 μm² area covered with SK QDs of an average width of 20 nm and a height of 3 nm. Both samples have a QD density of 1x10¹¹ cm⁻². The densities and sizes of both QD types can be varied by several orders of magnitude [6]. In our experiments we choose samples with similar QD densities to compare the luminescence behaviour.
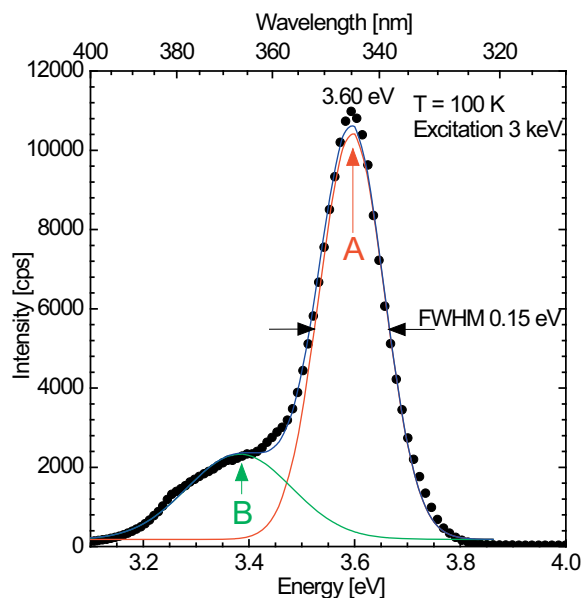

Figure 4 CL spectrum of c-GaN DE QDs with two Gaussian shaped emission bands peaking at 3.60 eV (A) and 3.38 eV (B). Emission band A with a FWHM of 0.15 eV is attributed to the ensemble luminescence of the QDs. Emission band B is related to the background luminescence.

Figure 5 CL spectrum of c-GaN SK QDs with two Gaussian shaped emission bands peaking at 3.58 eV (A) and 3.38 eV (B). Emission band A with a FWHM of 0.19 eV is attributed to the ensemble luminescence of the QDs. Emission band B is related to the background luminescence.
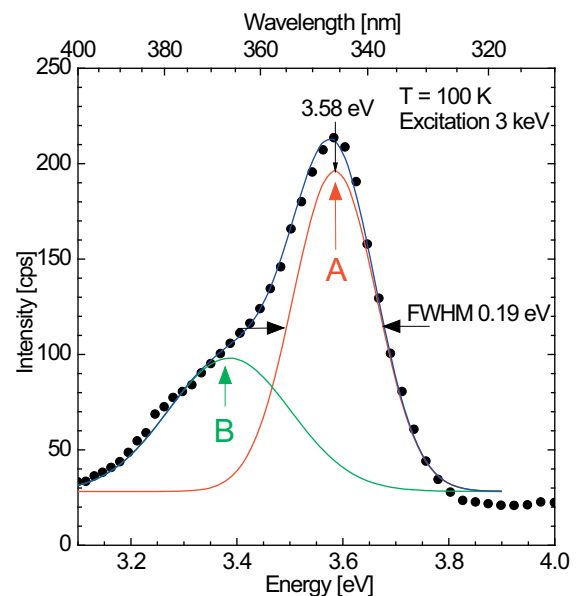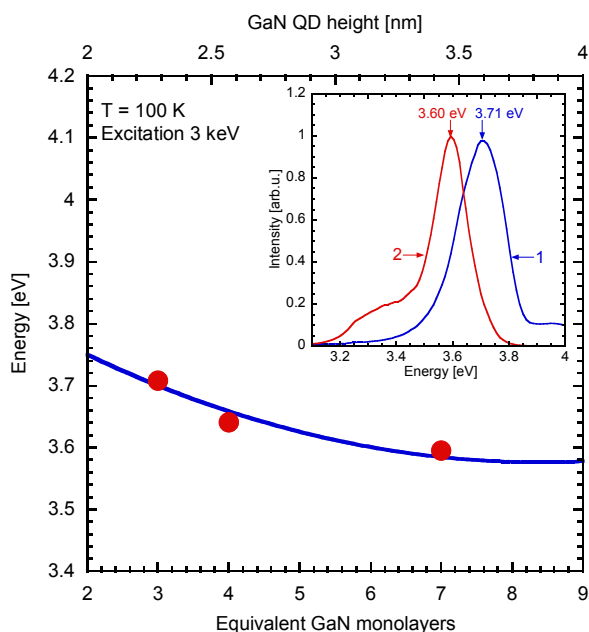
Figure 6 Correlation between QD emission energy of c-GaN DE QDs, deposited amount of GaN and calculated QD height. The experimental data are indicated by dots. The graph shows the calculated QD emission energy vs. QD height relation. The inset compares the energy shift of two CL spectra with different amount of deposited Ga.

3.3 Cathodoluminescence spectroscopy Compared to the broad CL spectrum of GaN bulk material, the emission of a single QD has a very narrow full width at half maximum (FWHM) idealized as a delta function. Therefore the ensemble luminescence is a superposition of many single QD emission lines. Since there are 10^5 QDs in the excitation area of 100 μm^2 the shape of the emission band is correlated to the size distribution of the QDs.

To compare the emission spectra of c-GaN DE QDs and SK QDs, two samples with a single layer of QDs and similar QD density were investigated. For all CL measurements an acceleration voltage of 3 keV was applied. Figure 4 shows a CL spectrum of DE QDs taken at 100 K. The CL spectrum consists of two Gaussian shaped emission bands peaking at 3.60 eV (A) and 3.38 eV (B). Due to the confinement energy of the QDs the transition energy is supposed to be much larger than the band gap energy of bulk c-GaN [13]. Therefore emission band A with a FWHM from 0.15 eV is related to the DE QDs luminescence. The value of 3.60 eV is in good agreement with the theoretical work of Fonoberov et al. [14].

Figure 5 shows the CL spectrum at 100 K of SK QDs. Two Gaussian shaped emission bands peaking at 3.58 eV (A) and 3.38 eV (B) are identified. Emission band A resembles the SK QDs ensemble luminescence. The origin of emission band B in Figs. 4 and 5 with a peak at 3.38 eV may be

attributed to an oil contamination by the vacuum pumps of our CL setup [15]. Emission band B is not observed in photoluminescence experiments [6].

In comparison to SK QDs the DE QDs luminescence intensity is higher by more than one order of magnitude.

As the QD emission energy is significantly dependent on the height of the QDs [14], a series of c-GaN DE QDs samples with different amounts of deposited Ga was investigated. Figure 6 shows the relationship between the QD emission energy and the amount of deposited Ga. The emission energy decreases in the range 3.71 eV-3.60 eV with increasing amount of deposited Ga from 3 to 7 ML, as indicated by dots. The inset illustrates exemplarily the energy shift of two CL spectra. Spectrum 1 corresponds to a sample with 3 ML and spectrum 2 to 7 ML of deposited Ga.

Calculations of the emission energy vs. QD height by V. Fonoberov et al. [14] are in good agreement with the experimental results, as shown in Fig. 6 (solid curve).

4 Conclusion We measured strong cathodoluminescence of cubic GaN QDs grown on 3C-AlN (001) by droplet epitaxy. For reference Stranski-Krastanov QDs were also investigated. The intensity of DE QDs luminescence is higher by more than one order of magnitude than SK QD luminescence at a comparable QD density. The size and therefore the emission energy of the DE QDs was varied by changing the amount of deposited Ga. The DE QD emission energy was tunable between 3.60 eV and 3.71 eV as measured by cathodoluminescence at low temperature (100 K). Finally, it was shown that the correlation of QD height and emission energy is in good agreement with calculations by Fonoberov et al. [14].

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