Growth of cubic GaN quantum dots

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Abstract. Zinc-blende GaN quantum dots were grown on 3C-AlN(001) by two different methods in a molecular beam epitaxy system. The quantum dots in method A were fabricated by the Stranski-Krastanov growth process. The quantum dots in method B were fabricated by droplet epitaxy, a vapor-liquid-solid process. The density of the quantum dots was controllable in a range of 10^8 cm^{-2} to 10^{12} cm^{-2}. Reflection high energy electron diffraction analysis confirmed the zinc-blende crystal structure of the QDs. Photoluminescence spectroscopy revealed the optical activity of the QDs, the emission energy was in agreement with the exciton ground state transition energy of theoretical calculations.

Keywords: Quantum dots, GaN, Stranski-Krastanov, droplet epitaxy, emission energy
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INTRODUCTION

Quantum optical devices based on wide-bandgap quantum dots (QDs) are of great interest for their possible application in quantum communication. For wurtzite GaN QDs, single photon emitters have been realized, showing photon antibunching and triggered single-photon generation.[1] However, the Quantum Confined Stark Effect caused by internal electric fields leads to a reduced recombination probability of electrons and holes in confined states. Consequently, the long radiative recombination time results in low repetition rate photonic devices. The metastable zinc-blende (cubic) phase of AlN (c-AlN) and GaN (c-GaN) has no polarization fields in (001) growth direction.[2] As a result, the radiative recombination time of c-GaN QDs was measured to be two orders of magnitude below the recombination time of wurtzite GaN QDs.[3] Up to now the method of choice to fabricate zinc-blende GaN QDs has been the Stranski-Krastanov (SK) growth process.[4]

In this contribution, we report on two different methods to fabricate c-GaN QDs. Method A comprises the fabrication of QDs by the Stranski-Krastanov (SK) growth process. These SK QDs are created by strain induced islanding in lattice-mismatched heteroepitaxy.[4] Method B is an alternative growth method for QDs by a vapor-liquid-solid process, namely droplet epitaxy (DE). In DE liquid Ga droplets are created and subsequently nitridated to fabricate GaN quantum dots.[5] DE has the advantage of size and density control of the QDs over a wide range from 10^8 cm^{-2} to 10^{12} cm^{-2}. For applications as a single photon emitter the low density QDs are of great importance, because the minimal spatial separation of the QDs has to be in the order of the emission wavelength.

EXPERIMENT & DISCUSSION

The growth of the c-GaN QDs and the c-AlN buffer was carried out in a Riber 32 MBE system, including an Oxford Instruments N plasma cell and standard Ga and Al effusion cells. Reflection high energy electron diffraction (RHEED) was available for in-situ growth monitoring. Atomic force microscopy (AFM) was utilized to analyze the surface structure of the grown layers and QDs. Photoluminescence (PL) spectroscopy was applied to obtain optical data. The samples were excited by an ArF excimer laser with λ=193nm. The PL was detected by a liquid nitrogen cooled charge coupled device camera mounted to a grating monochromator with a focal length of f=1 m. The samples were mounted in a temperature controllable liquid helium cryostat to allow for PL investigations at different temperatures.

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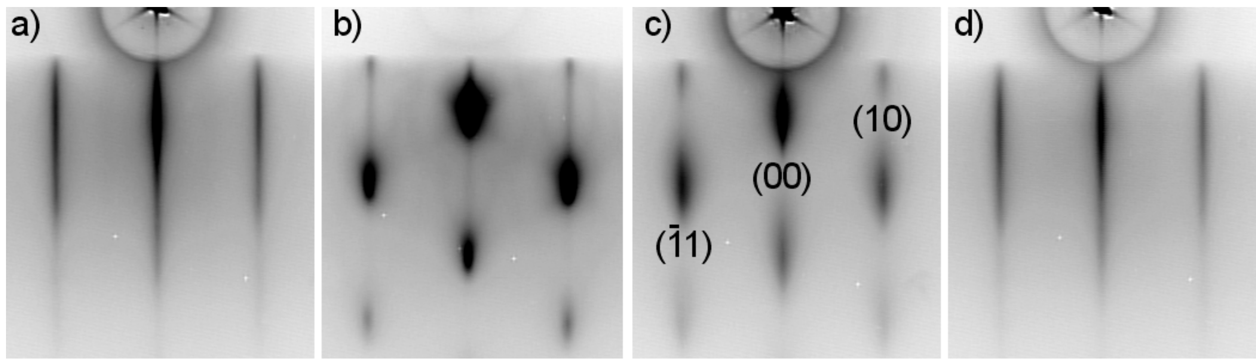

FIGURE 1. RHEED patterns: (a) Reflections of the c-AlN layer, long thin streaks indicate a smooth 2D surface. (b) Reflections of c-GaN SK QDs, the spotty reflections indicate 3D islands of cubic crystal structure. (c) Reflections of c-GaN droplet QDs. (d) Reflections of the c-AlN after 30 ML QD overgrowth.

For both c-GaN QD growth methods, the first step is to grow the lower confinement layer. A 30 nm c-AlN (001) layer is deposited on 3C-SiC (001) substrate at 730° C substrate temperature by plasma assisted molecular beam epitaxy (PAMBE).[6] The RHEED pattern of the AlN surface in [110] azimuth, shown in Fig. 1(a), has long thin streaks, including streaks of a twofold surface reconstruction. These streaks indicate a smooth two-dimensional (2D) surface.[7] The atomically smooth AlN surface is verified by AFM.

The driving force in a SK process is the strain energy.[4] For c-GaN the lattice misfit between 3C-SiC (a=4.36 Å) and c-GaN (a=4.50 Å) is 3.2%.[8,9] Cubic AlN is pseudomorphically strained on 3C-SiC and has a lateral lattice parameter of a=4.36 Å. When growing c-GaN the initial GaN monolayers (ML) are pseudomorphically strained on c-AlN. Once the critical layer thickness of 2 ML is reached, SK GaN QDs are formed to relief the strain. For the fabrication of SK GaN QDs the equivalent of 2 to 10 ML GaN are deposited on the AlN layer. Figure 1(b) shows the RHEED pattern after the formation of the GaN SK QDs. The spotty reflections are a result of a transmission electron diffraction component through three dimensional islands on the surface.[7] The spotty reflections are therefore an indication of quantum dots on the surface. The broadening of the reflections can be explained by a change in lattice parameter of the upper part of the QDs due to partial relaxation.

In droplet epitaxy liquid Ga droplets are created and subsequently nitridated to fabricate GaN QDs. Initially the equivalent of 2 to 5 ML of Ga is deposited on the AlN surface at 350 °C substrate temperature. The formation of droplets is induced by the strong cohesion force between the Ga atoms. The combination of the amount of deposited Ga and the substrate temperature can be used to control the size and density of the droplets.[5] In the next step the Ga droplets are exposed to the N plasma beam for 3 to 10 minutes. During the nitridation the substrate temperature is gradually ramped up from 350° C to 730° C. Figure 1(c) shows the RHEED pattern after the nitridation of the Ga droplets. Similar to the case of SK QDs spotty reflections indicate quantum dots on the surface. All reflections in the RHEED pattern indicate zinc-blende crystal structure. Reflections of the wurtzite lattice would be alongside the diagonal of the (-11) reflection to the (10) reflection, but are absent.[10]

We determined the size of the QDs and the distribution on the surface by AFM of uncapped GaN QDs. Figure

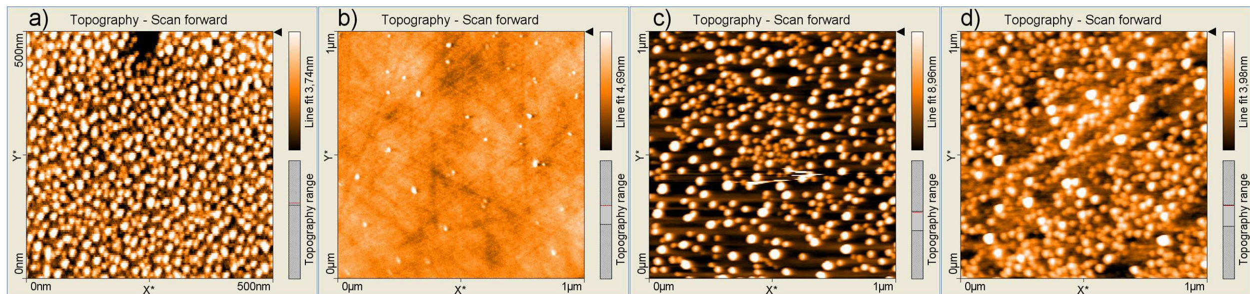

FIGURE 2. AFM images: (a) 0.5x0.5 μm^2 area of GaN SK QDs on c-AlN, density $5*10^{11}$ cm^{-2}. (b) 1x1 μm^2 area of GaN DE QDs on c-AlN, density $3*10^9$ cm^{-2}. (c) 1x1 μm^2 area of GaN DE QDs on c-AlN, density of large QDs $3*10^{10}$ cm^{-2}, density of small QDs $6*10^{10}$ cm^{-2}. (d) 1x1 μm^2 area of GaN DE QDs on c-AlN, density $1*10^{11}$ cm^{-2}.

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2(a) shows an AFM image of a 0.5x0.5 µm^2 area of the AlN surface covered with SK GaN QDs. The average width of the QDs is 15 nm, the height 3 nm and the density $5*10^{11}$ cm^{-2}. The high density is typical for SK QDs, as SK QDs can only be formed after the critical thickness amount of GaN has been deposited. However, for DE the limitation to high densities does not apply, as shown in the AFM image in Fig. 2(b). The average width of the QDs is 10 nm, the height 3 nm and the density $3*10^{9}$ cm^{-2}. Figure 2(c) shows an AFM image of GaN DE QDs of medium density. A bimodal size distribution can be observed. The average width of the larger QDs is 30 nm, the height 6 nm and the density $3*10^{10}$ cm^{-2}. The average width of the smaller QDs is 15 nm, the height 4 nm and the density $6*10^{10}$ cm^{-2}. The bimodal size distribution can be explained by Ostwald ripening. Large QDs feed on neighboring small QDs, with the result of larger, but more distant QDs.[11] This can be observed in the lower left quadrant of Fig. 2(c), there the concentration of large QDs is higher and the concentration of small QDs is lower than average. Last but not least Fig. 2(d) shows an AFM image of GaN DE QDs of high density. The average width of the QDs is 20 nm, the height 3 nm and the density $1*10^{11}$ cm^{-2}. Compared to SK QDs the size distribution is significantly larger. Our AFM studies revealed, that Ostwald ripening can be utilized to modify size and density of the GaN DE QDs.

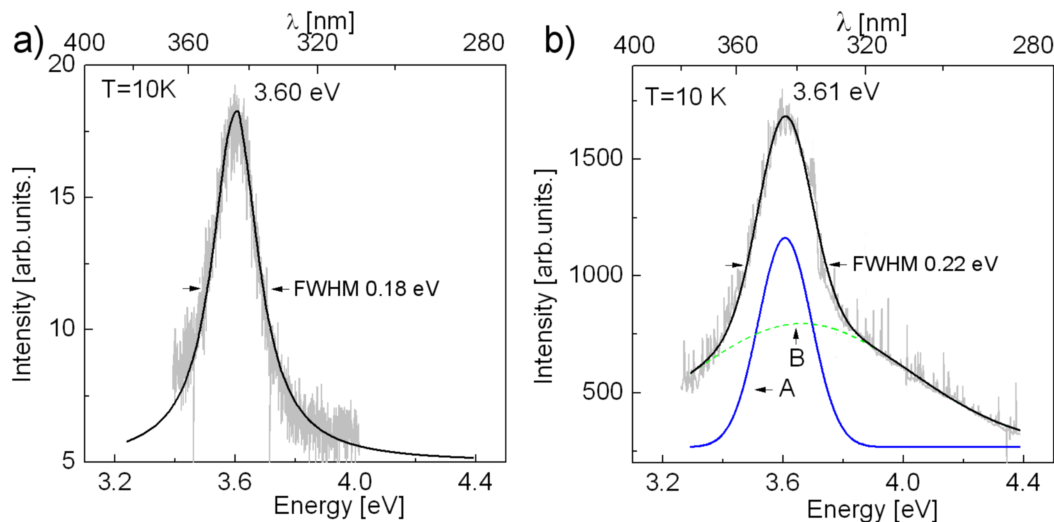

FIGURE 3. Photoluminescence spectra:
(a) Gaussian shaped emisson band of c-GaN SK QDs peaking at an energy of 3.60 eV.
(b) Two overlapping Gaussian shaped emisson bands of c-GaN droplet QDs, one at an energy of 3.61 eV ±0.01 eV, marked with A, and one at 3.64 eV ±0.01 eV, marked with B.

For both c-GaN QD growth methods, the final step is the overgrowth of the QDs with the upper confinement layer of AlN. Figure 1(d) shows the RHEED pattern of the AlN surface after 30 ML of QD overgrowth. Long thin streaks indicate a smooth 2D surface, spotty reflections are no longer present, full epitaxial overgrowth of the GaN QDs can be concluded. AFM measurements of the AlN surface after GaN QD overgrowth confirm a smooth surface.

To analyze the photoluminescence of the c-GaN QDs, comparable samples with and without 30 nm AlN cap layer were grown. Overview PL spectra were taken in the energy range from 3 eV to 6 eV, but as no PL luminescence was observed above 4.5 eV, the high resolution spectra were measured up to 4.5 eV. Figure 3(a) shows the PL spectrum of a c-GaN SK QDs sample at 10 K. The amount of deposited GaN equals 8 ML. A Gaussian shaped emission band peaking at an energy of 3.60 eV ±0.01 eV can be identified. Since the QD transition energy is mainly influenced by the QD height, AFM is utilized to determine the QD height. The average height of the uncapped QDs is 3.3 nm ±0.2 nm. The theoretical work of Fonoberov et al.[12] on c-GaN QDs predicts an exciton ground state transition energy of 3.59 eV for QDs having a dot height of 3.3 nm above the wetting layer. This energy is in good agreement with the measured energy peak at 3.60 eV of the capped c-GaN SK QDs. Figure 3(b) shows the 10 K PL spectrum of a c-GaN DE QDs sample. The amount of deposited GaN equals 5 ML. Two overlapping Gaussian shaped emission bands are identified, one at an energy of 3.61 eV ±0.01 eV, marked with A, and a broader one at 3.64 eV ±0.01 eV, marked with B. The average height of the uncapped DE QDs is 3.1 nm ±0.2 nm. The calculations predict an exciton ground state transition energy of 3.62 eV for QDs having a dot height of 3.1 nm

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above the wetting layer. This energy is in good agreement with the measured energy peak at 3.61 eV of the capped c-GaN DE QDs. The good agreement underlines the existence of a wetting layer. The existence of two overlapping emission bands can be explained by partial Ostwald ripening. The initially formed QDs are smaller and have a larger size distribution, the emission maximum is at an energy of 3.64 eV. The ripened QD are bigger and have a smaller size distribution, consequently the emission maximum is at an energy of 3.61 eV.[5] Spectra of other c-GaN DE QDs however, show only single emission bands. To exclude other possible origins of the emission, the exciton ground state transition energies for various c-GaN quantum wells (QW) strained on c-AlN are calculated by solving the Schrödinger equation. The calculated exciton ground state transition energy for a 5 ML c-GaN QW is 4.05 eV +- 0.025 eV, thinner QWs have even higher transition energies. The energy of c-plane wurtzite GaN QDs of the same size is 3.20 eV.[12] As a result, c-GaN QWs and wurtzite GaN QDs can be ruled out as origin for the 3.61 eV emission, confirming the c-GaN QDs as the emission source.

CONCLUSIONS

Zinc-blende GaN quantum dots were grown on 3C-AlN(001) by two different methods. The Stranski-Krastanov growth process yielded c-GaN QDs of high density and narrow size distribution. The PL spectrum of the SK QDs showed a narrow emission band at 3.60 eV. The droplet epitaxy method allowed to grow c-GaN QDs in a wide range of densities down to 10^8 cm^{-2}. The PL spectrum of the droplet epitaxy QDs showed two overlapping emission bands at an energy of 3.61 eV and 3.64 eV, respectively. The determined energies were in good agreement with the calculated exciton ground state transition energies of c-GaN QDs. In conclusion, we have shown that droplet epitaxy can be utilized to create optically active zinc-blende GaN QDs.

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