ARTICLE IN PRESS

Nuclear Instruments and Methods in Physics Research B xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

# Ion-induced interdiffusion of surface GaN quantum dots

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A R T I C L E I N F O

*Article history:*
Received 5 December 2016
Received in revised form 7 April 2017
Accepted 11 April 2017
Available online xxxx

*Keywords:*
GaN/AlN
Quantum dots
Ion beam implantation
Cathodoluminescence
Quantum confined Stark effect

A B S T R A C T

In the flourishing fields of quantum technology gallium nitride (GaN) quantum dots (QDs) have great appeal by providing high stability and room-temperature operation. Here, we report on the ion implantation of surface GaN QDs grown in the hexagonal crystal structure. An uncapped sample (S1) and two samples capped by 8 ML (S2) and 16 ML (S3) of AlN are subjected to a 100 keV gallium (S1, S2) and a 210 keV erbium (S3) ion beam. The fluence ranged from $5 \times 10^{10}$ cm$^{-2}$ to $1 \times 10^{15}$ cm$^{-2}$ (S1, S2) and from $5 \times 10^{10}$ cm$^{-2}$ to $5 \times 10^{13}$ cm$^{-2}$ (S3). QD characterization is performed by cathodoluminescence measurements at 77 K and atomic force microscopy and scanning electron microscopy.

Strong interdiffusion processes upon ion impact at the interfaces are evidenced leading besides other effects to a quenching of the quantum confined Stark effect. Moreover, a model for the QD morphology based on a fluence-dependent diffusion coefficient is developed.



## 1. Introduction

Since the last two decades, buried hexagonal GaN QDs have been subject to extensive research being high efficient light emitters [1]. Fundamental interest exists especially for the present huge piezoelectric fields affecting the luminescent properties. These fields lead to a strong quantum confined Stark effect (QCSE) which results in a band bending with a spatial separation of electron and hole wavefunction. On the one hand, the radiative recombination is reduced, on the other hand a strong redshift of the QD emission to an energy even below the GaN bandgap occurs [2]. By adjusting further fabrication parameters, the emission wavelength can be tuned over a wide spectral range from ultraviolet to the orange range [3]. Moreover, unique properties like a large exciton binding energy and a strong quantum confinement enable room-temperature operation. These promising features make them highly attractive as single-photon sources [4,5] for quantum communication technologies and spin-electronic devices for quantum computing [6], raising the demand on single electrically and optically active QDs. One approach for the realization of those could be a top-down process using focused ion beam implantation to post-select self-assembled molecular beam epitaxy-grown QDs [7] by introducing non-radiative defects around an intentional QD. Further, magnetic impurities could be incorporated by rare earth implantation with low fluences in the non-disabled QDs for spin control. This ion beam engineering implies the necessity of a strong expertise of the implantation effects. Investigations on surface QDs allow to go beyond the experimental opportunities applied to deeply buried QDs. Here, we study the ion-induced effects on surface hexagonal GaN QDs which offers a direct access to the affected QD morphology.

Besides these advantages for fundamental research, surface QDs also have a high potential in sensor applications as they are very sensitive to environmental changes [8].

This work is organized as following: In Section 2 the experimental procedure including QD growth, ion implantation and QD characterization is described. In Section 3.1 fluence-dependent cathodoluminescence (CL) spectroscopy on uncapped and capped GaN QDs is discussed. Afterwards, atomic force microscopy (AFM) and scanning electron microscopy (SEM) measurements on the surface QDs are analyzed in Section 3.2, followed by concluding remarks in Section 4.

## 2. Materials and methods

Three samples with each a single layer of GaN QDs were grown in the hexagonal (wurtzite) crystal structure using molecular beam epitaxy. The layer sequence of the samples consists of a Si (111) substrate, covered with about 370nm of AlN. On top, the QDs

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http://dx.doi.org/10.1016/j.nimb.2017.04.036
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formed in the Stranski–Krastanov growth mode. Here, samples with different QD capping were investigated. The first sample (S1) was uncapped, the second (S2) and third sample (S3) were capped by 8ML and 16ML of AlN, respectively. The QDs with a bottom length of around 9 nm and a height of (3–4) nm are grown in densities of $2 \cdot 10^{11}$ cm$^{-2}$ (S1), $1.2 \cdot 10^{11}$ cm$^{-2}$ (S2) and $0.8 \cdot 10^{11}$ cm$^{-2}$ (S3).

The samples were implanted in an EIKO-100 FIB system with a limitation to acceleration voltages between 70 kV and 100 kV. For providing comparable penetration depths, the ions were implanted with energies of 100 keV for single charged gallium ions (S1, S2) and 210 keV for erbium ions in the charge state of $3^+$ (S3). To reach the highest damage inside the QD layer, the samples were covered by 100 nm of PMMA before implantation. Inside this PMMA/GaN/AlN/Si system, the gallium ion distribution reaches its maximum in a depth of 111 nm with a straggle of 15 nm and the erbium ions reach an average depth of 118 nm with a straggle of 10 nm, according to simulations with the SRIM software [9]. Further, a number of total displacements of around 2.4 (Å·ion)$^{-1}$ in the QD layer for the gallium ion bombardement and of about 5.0 (Å·ion)$^{-1}$ for the erbium ion irradiation is estimated by SRIM simulations. Fields of $(1 \times 1)$ mm$^2$ of size were scanned with the ion beam in a fluence range from $5 \times 10^{10}$ cm$^{-2}$ to $1 \times 10^{15}$ cm$^{-2}$ (S1, S2) and from $5 \times 10^{10}$ cm$^{-2}$ to $5 \times 10^{13}$ cm$^{-2}$ (S3).

After removing the PMMA, the samples were optically characterized by cathodoluminescence spectroscopy using a 5keV electron beam with currents of $7 \cdot 10^{-10}$ A scanning an area of about $(55 \times 55)$ μ m$^2$ of size. The signal was detected at 77 K by a CCD camera. The QD morphology was analyzed by atomic force microscopy (AFM) and scanning electron microscopy (SEM) concerning their density and size with respect to the ion fluence.

## 3. Results

### 3.1. Fluence-dependent CL spectroscopy

Figs. 1(a)–(c) show CL spectra measured at 77 K of surface QD ensembles irradiated with different ion fluences. A luminescence quenching can be observed with increasing ion fluence, which is analyzed in detail by the fluence-dependent CL intensity plotted in Fig. 1(d). This quenching has been already extensively studied for deeply buried InAs/GaAs QDs [10,11] and was also observed for deeply buried hexagonal GaN/AlN QDs [12]. The fluence-dependent integrated intensity of the latter was described by a model accounting not only for an increase of the non-radiative recombination rate due to ion-induced non-radiative defect creation but also for an increase of the radiative recombination rate [12]. Its increase was suggested to be due to intermixing processes resulting in smeared out QD profiles with compositional changes and thus in a quenching of the QCSE [13].

Here, the experimental integrated intensity follows the same model as found for the deeply buried QDs, as illustrated in Fig. 1 (d). Thus, the surface QDs exhibit comparable behavior to deeply buried QDs under ion bombardment. Moreover, they also show a strong degradation resistance as reported in [12] considering an average number of 100 implanted ions per QD for the highest ion fluence of $10^{14}$ cm$^{-2}$, for which CL still could be observed.

Besides the intensity drop, also a CL blueshift relative to the unimplanted sample is present in the lower fluence range, whereas at higher fluences above $10^{12}$ cm$^{-2}$ a CL redshift dominates. This behavior is ascribed to the reduction of the QCSE for low fluences resulting in an increase of the QD emission energy [14] and to the competing effect of a strain-induced energy decrease for the higher fluences [7]. Thus, at a critical damage threshold, the QCSE itself and its reduction just weakly influence the QD luminescence.

To gain further insight, CL measurements at different electron beam exposure times on the uncapped sample S1 are performed. First, it is worth to note that the QD peaks in Fig. 1 originate from QDs with different heights where smaller QDs emit at higher energies [15]. Moreover, higher QDs are more strongly affected by the QCSE due to the larger influence of the internal electric field $F$ on the emission energy $E$ according to $E = c - eFh$, with the constant $c$ and the height $h$ [16].

In Fig. 2, the evolution of the QD emission energies with increasing exposure time $t$ of the unirradiated QD ensemble is as expected. The emission energy around 2.92 eV at $t = 0$ s for the higher QDs increases (Fig. 2(a)), as the electron irradiation causes a charge accumulation with a partial screening of the internal electric field and a reduction of the QCSE [2]. The emission energy of around 3.52 eV at $t = 0$ s for the smaller QDs nearly stays constant (Fig. 2(b)). The latter is also true for the smaller irradiated QDs, but the rise of the CL energy of the irradiated high QDs is weaker. This confirms the afore assumed reduction of the QCSE in the implanted QD samples.

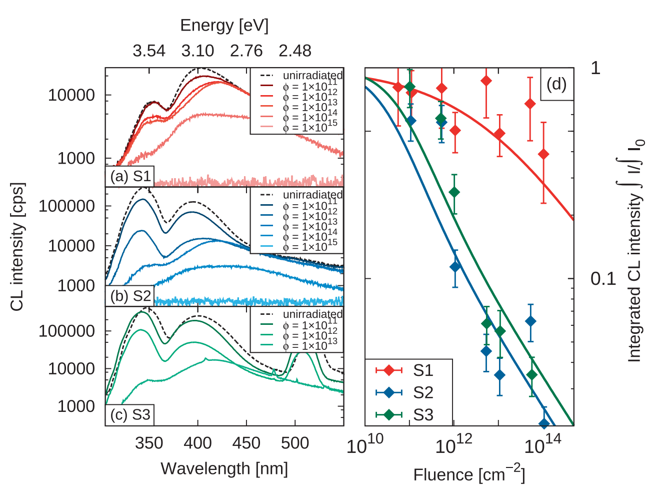


**Fig. 1.** CL spectra measured at 77 K for the ion irradiated surface QDs with different capping. The peaks above 2.9 eV are attributed to QD luminescence with an underlying broad defect peak located at lower energies, see also [16,12]. In (c) a second order peak around 515 nm related to the GaN wetting layer is present. Fluence values are given in units of cm$^{-2}$. In (d) the fluence-dependent integrated CL intensity is shown, fitted with the model (solid lines) from [12].

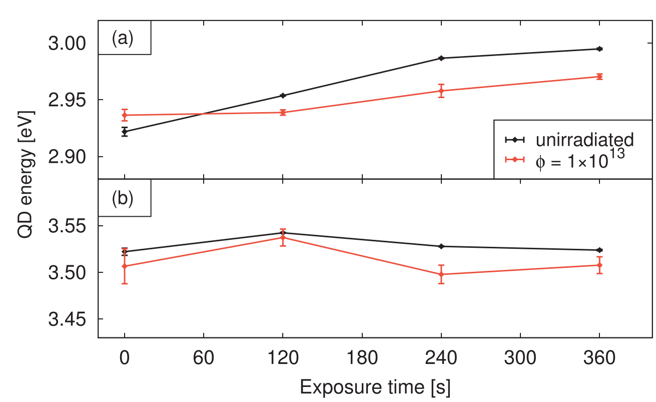


**Fig. 2.** QD emission energy for unirradiated and irradiated QD ensembles measured by CL at 77 K at different electron beam exposure times in a scan area of $(11 \times 11)$ μ m$^2$ of size using an electron beam current of $5 \cdot 10^{-11}$ A. Emission energies below 3.0 eV (a) and above 3.5 eV (b) of ensemble QDs are shown. Fluence values are given in units of cm$^{-2}$.