Intramolecular electron transitions in GaN/AlN quantum dot superlattices

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We report the Stranski–Krastanov growth of Si-doped GaN/AlN quantum dot superlattices displaying conduction-band intraband absorption at telecommunication wavelengths. By adjusting the growth conditions, quantum dots with a height of 0.5–1 nm in the density range of 0.1 \times 10^{12}–3 \times 10^{12} \text{ cm}^{-2} can be synthesized. All of the samples exhibit room-temperature intraband absorptions in the 1.41–1.54 \mu\text{m} wavelength range. The full width at half maximum of the observed intraband absorptions is as small as 88 meV. We also observe residual intraband absorption for nominally undoped samples. We attribute this effect to the detrapping of electrons in the AlN barriers and subsequent capture in the dots. © 2005 American Institute of Physics.

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TABLE I. Sample growth parameters (doping level and GI time), QD heights (above the 0.5-nm-thick wetting layer), diameter and density estimated from AFM measurements, experimental PL energy and FWHM, intraband peak energy, FWHM, and absorption per reflection. All of the measurements have been performed at room temperature.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Doping (cm⁻³)</th>
<th>GI time</th>
<th>QD height (diameter) (nm)</th>
<th>QD density (cm⁻³)</th>
<th>Intraband energy (FWHM) (eV)</th>
<th>Absorption per reflection (%)</th>
<th>PL (FWHM) (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E645</td>
<td>n.i.d.</td>
<td>15 s</td>
<td>1.3±0.6(11±2)</td>
<td>1.2±0.1×10¹²</td>
<td>0.823 (0.094)</td>
<td>1.4</td>
<td>3.826 (0.195)</td>
</tr>
<tr>
<td>E646</td>
<td>1.1×10¹⁹</td>
<td>15 s</td>
<td>1.3±0.6(10±2)</td>
<td>1.1±0.1×10¹²</td>
<td>0.826 (0.094)</td>
<td>2.4</td>
<td>3.824 (0.195)</td>
</tr>
<tr>
<td>E644</td>
<td>1×10²⁰</td>
<td>15 s</td>
<td>1.3±0.6(11±5)</td>
<td>1.2±0.6×10¹⁰</td>
<td>0.837 (0.112)</td>
<td>5.1</td>
<td>3.818 (0.261)</td>
</tr>
<tr>
<td>E649</td>
<td>1×10²⁰</td>
<td>0 s</td>
<td>0.9±0.4(11±2)</td>
<td>2.9±0.1×10¹²</td>
<td>0.878 (0.088)</td>
<td>3.1</td>
<td>3.894 (0.246)</td>
</tr>
<tr>
<td>E647</td>
<td>1×10²⁰</td>
<td>1 min</td>
<td>1.2±0.6(20±3)</td>
<td>4±0.4×10¹¹</td>
<td>0.823 (0.1)</td>
<td>3.8</td>
<td>3.82 (0.24)</td>
</tr>
<tr>
<td>E648</td>
<td>1×10²⁰</td>
<td>2 min</td>
<td>1.5±0.7(28±2)</td>
<td>1±0.2×10¹¹</td>
<td>0.81 (0.125)</td>
<td>5.2</td>
<td>3.816 (0.237)</td>
</tr>
</tbody>
</table>

their diameter. This process is known as “Ostwald ripening”. Indeed, the data in Table I show how the QD diameter evolves from 10±3 nm to 30±5 nm by increasing the GI time from 0 s to 2 min. It is important to notice that, for this material system, the AFM is a good probe of morphological characteristics of the QDs, since the AlN capping modifies the strain state of the QDs, but it does not significantly alter the QD shape.¹¹ To guarantee that the GaN islands on the surface are not subject to additional ripening, the samples were rapidly quenched to room temperature immediately after the GI that follows the deposition of the last QD layer.

Photoluminescence (PL) experiments were carried out at room temperature using the UV excitation line (λ=244 nm) of a frequency-doubled continuous-wave Ar⁺ laser. The PL signal was focused into a 0.46 m focal length monochromator, and detected with a charge coupled device camera. The PL peak energies, along with the corresponding FWHM, are reported in Table I. For the sample grown with shorter GI, the PL peak energies, along with the corresponding FWHM, are in agreement with the smaller QD size measured by AFM. Inset: Room-temperature PL spectra for Samples E644, E648, and E649. The PL is peaked at energies ranging from 3.78 to 3.9 eV, which is typical of dots with a height of 4–5 MLs (1–1.3 nm).¹³ The FWHM is in the range of 0.2–0.25 eV. This value is remarkably small for nitride QDs, and it suggests a good size homogeneity.

For the infrared (IR) absorption measurements, the samples were mechanically polished in a 45° multipass waveguide geometry with 4–5 total internal reflections. The IR transmission for p- and s-polarized light was measured at room temperature using a Fourier transform IR spectrometer and a deuterated triglycine sulfate photodetector. Figure 2 shows the absorption of Samples E644 and E649 for p polarization and of Sample E648 for s polarization. These QD samples exhibit p-polarized absorption peaked between 0.81 eV (1.53 μm) and 0.878 eV (1.41 μm). As seen in Fig. 2, no absorption is observed for s-polarized light within experimental accuracy. The oscillating feature arises from Fabry–Perot interferences in the epitaxial layers and AlN buffer. The p-polarized interlevel absorption energy of all studied samples, along with the corresponding FWHM, is reported in Table I. For the sample grown with shorter GI, the interlevel transition is peaked at higher energies. This is in agreement with the smaller QD size measured by AFM. The FWHM is as low as 88 meV for Sample E649.

FIG. 1. AFM image of QDs on the surface of Sample E644 obtained in tapping mode using a Dimension 3100 system.

FIG. 2. Room-temperature intraband absorption of Samples E644 (full line with triangles), E648 (full line with squares), and E649 (full line with circles) for p-polarized light and of Sample E648 (dashed line) for s-polarized light. The measurements were performed in a multipass waveguide configuration with four internal reflections. Inset: Room-temperature PL spectra for Samples E644, E648, and E649.
our previous findings,\textsuperscript{6} we attribute the $p$-polarized peak to the transition from the ground state ($s$) to the excited state confined along the $c$ axis ($p_z$). The fundamental absorption of the dots from the ground state $s$ to the in-plane excited states ($p_x$ or $p_y$) is not observed because it probably occurs below the cut-off energy of the substrate,\textsuperscript{14,7} located at 0.3 eV ($\lambda = 4.1 \mu m$). However, one cannot exclude that the $s-p_z$ and $s-p_x$ absorption for heavily doped samples is vanishingly small due to the excited state filling.

The thickness and length of the multipass waveguides was set to allow four to five internal reflections. Under these conditions, the samples that were $n$ doped to $1 \times 10^{20} \text{cm}^{-3}$ exhibit an absorption in the 12.4–20.8\% range. For comparison purposes, the per-reflection absorption (i.e., the measured absorption divided by the number of total internal reflections) is summarized in Table I for all the investigated samples. Although the absorption increases with doping, its trend is not linear. In addition, Sample E645—which is nominally undoped—shows a weak $p$-polarized absorption peaked at 0.823 eV (1.51 $\mu m$) with a FWHM of 94 meV (see Fig. 3, inset).

The latter unexpected finding cannot be explained by the GaN residual doping, which is estimated to be $\sim 10^{17} \text{cm}^{-3}$ and, therefore, is too weak to provide a detectable electronic population. One possible explanation is the efficient trapping of electrons in the dots from impurity states located in the AlN barriers. It is well known that oxygen is a common substitutional impurity which can be present in large concentrations in AlN. The incorporation of oxygen in AlN leads to a deep-defect band with a broad energy range from $\sim 0.7 \text{ eV}$ to $\sim 2.7 \text{ eV}$ below the conduction band of AlN.\textsuperscript{15} It is expected that the deep levels—with an energy higher than that of the QD ground state, typically 1 eV below the AlN conduction band—are responsible for the population of the dots. Deeper levels can also be activated for example using light illumination. Indeed, we have observed that the interlevel absorption of Sample E645 (and of all the doped samples, too) increases under illumination by either a UV or a visible argon laser. Furthermore, after illumination, the interlevel absorption returns to its initial value of 6.2\% within a few minutes. Figure 3 shows a typical absorption decay after sample illumination. A decay time constant of $\sim 43 \text{ s}$ is estimated using the exponential fit. Such a long recovery time is in agreement with a simplified model where electrons photoexcited from deep defects in the AlN barriers are then captured by the QDs. The extremely long lifetime stems from the spatial separation between the donors, located in the AlN barriers, and the electrons efficiently trapped in the QDs. We stress that the unintentional doping of the AlN layer is difficult to control and this fact may explain the observed dispersion of interlevel absorptions for the doped samples.

In conclusion, we have reported the growth of high-density Si-doped self-organized GaN/AlN QDs displaying conduction-band interlevel absorptions at telecommunication wavelengths. The Si-doped QD absorption is shown to be dependent on the growth interruption time. Nominally undoped samples show residual intraband absorption as well as light-activated absorption, which are attributed to the detraping of electrons in the AlN barriers and subsequent capture in the dots.

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