THz absorbing AlGaN/GaN multi-quantum-wells: Demonstration of a robust 4-layer design

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Abstract

We report on AlGaN/GaN multi-quantum-well structures displaying intersubband absorption in the THz spectral range. Firstly, we theoretically analyze the weaknesses of the state-of-the-art GaN-based step-quantum-well architecture from an optoelectronic standpoint. We then propose a modified geometry with improved structural robustness considering the uncertainties associated to the growth. This later structure, consisting of 4-layer quantum wells, has been grown by plasma-assisted molecular-beam epitaxy, and characterized structurally and optically. Low temperature absorption of samples with different Si doping levels confirms intersubband transitions in the far-infrared, centred at 28 µm.
Wide-bandgap AlGaN/GaN heterostructures are intensively studied as a promising alternative to replace GaAs as the dominant material in solid-state THz optoelectronics. GaAs-based quantum cascade lasers operating in this spectral range are limited by intrinsic material properties, namely the LO phonon, which exists at 36 meV (34 µm). On the contrary, the large energy of GaN longitudinal-optical phonons (92 meV, 13 µm) opens prospects for GaN-based high-temperature THz quantum cascade lasers [1-5] and other intersubband (ISB) devices covering the 5-10 THz band, inaccessible to As-based technologies.

ISB transitions in GaN/AlGaN multi-quantum-wells (MQWs) can be tuned from 1.0 µm to 10 µm by reducing the Al mole fraction in the barriers and increasing the quantum well (QW) width [6-12] (see ref. [13] for a review). This strategy faces a roadblock at longer wavelengths because of the quantum confined Stark effect (QCSE) present in these polar materials. The polarization-induced internal electric field results in QWs with a triangular profile, and the lower energy electronic levels get confined into a small fraction of the QW, so that changes in well width do not induce proportional changes in ISB energy. Therefore, to shift the absorption towards longer wavelengths, it is necessary to engineer the conduction band to reduce the internal electric field within the QWs. This has led to a 3-layer step-QW architecture, aimed to flatten the band structure of the QW [14]. Following this design, ISB transitions in the THz region have been reported [14], and a QW infrared photodetector has been demonstrated [15]. However, this design suffers from issues with reproducibility and the inability to easily predict the ISB transitional energy. To progress towards more complex and performing devices, it is therefore important to introduce a different architecture with a higher robustness with regard to the growth uncertainties.

In this work, we discuss the properties of AlGaN-based MQWs designed for ISB
optoelectronics in the THz spectral range. We analyse the reproducibility issues associated to the current step-QW architecture and we propose an alternative, robust design based on a 4-layer QW. We demonstrate that this structure, fabricated by plasma-assisted molecular-beam epitaxy (PAMBE), presents TM-polarized ISB absorption at 26 µm, with magnitude and line width that increases with the doping level.

The experimentally observed reproducibility problems associated to the step-QW geometry were analyzed theoretically using the self-consistent nextnano³ 8-band k·p Schrödinger-Poisson solver [16]. Calculations were performed using the material parameters in ref. [17], neglecting all the bowing parameters for AlGaN, and assuming the structure strained on GaN. Figure 1 illustrates the electronic profile of an Al₀.₀₅Ga₀.₉₅N/Al₀.₁Ga₀.₉N/GaN (10 nm / 3 nm / 3 nm) step-QW, which presents an energy difference between the first and the second electronic levels of 35 meV (35.6 µm), a significant deviation from the measured value of 17.5 meV (70 µm) [14]. The robustness of this system with respect to the uncertainties associated to PAMBE growth has been analyzed in Figure 1. There are 5 degrees of freedom within the structure, namely the thickness of each layer and the Al mole fraction in each AlₓGa₁₋ₓN layer. Each degree of freedom was varied considering realistic deviations in the epitaxial growth: ±2 monolayers (ML) as the error bar for thicknesses and ±10% as the error bar for the aluminium mole fraction in AlₓGa₁₋ₓN, which corresponds to a temperature difference of ±4°C for the Al effusion cells in the compositional range of this design. While some parameters evoke little variance, errors induced upon the GaN well width and the Al concentration in the step-QW induce a drastic change in ISB wavelength, which can reach up to 40% variance, from 35 µm to 50 µm.

This 3-layer structure is designed around the principle of band edge equivalency. The step-QW can be broken effectively into two portions; the first is the “barrier”, which
comprises of the high-Al-content Al\textsubscript{x}Ga\textsubscript{1-x}N layer and the GaN layer. The second portion is the well, which is the low-Al-content Al\textsubscript{x}Ga\textsubscript{1-x}N layer. Having the “barrier” balanced at the same average Al percentage as the well ensures semi-flat band conditions in the well. This configuration is associated to the minimum energy spacing between the ground electronic state and the first excited state, as described in ref. [18]. The weakness of this design lies in the fact that any deviation from this balance results in an internal electric field in the well, which shifts the wavefunction associated to the first electronic level towards the GaN layer (Fig. 1(b) top, for higher Al content in the well) or towards the high-Al-content layer (Fig. 1(b) down, for lower average Al content in the well). Thus, any imbalance in the structure has a drastic effect on the ISB wavelength.

To surmount the limitations of the step-QW configuration, we propose a design which includes the insertion of an additional AlGaN layer to separate the GaN layer from the low-Al-content Al\textsubscript{x}Ga\textsubscript{1-x}N well. The “separation layer” is designed so that there is no confined state in the GaN layer. This architecture, described in Fig. 2(a), does not evade the quantum confined Stark effect, but the GaN layer contributes to reduce the average spontaneous polarization of the complex barrier structure (Al\textsubscript{0.1}Ga\textsubscript{0.9}N/GaN/ Al\textsubscript{0.07}Ga\textsubscript{0.93}N), which results in a lower electric field in the QW. The robustness of this design is analyzed in Fig. 2(c) and shows much less variation of the ISB transition energy than the step-QW architecture with respect to the growth uncertainties. The “separation layer” also has an inherent robustness on the order of the rest of the structure and should not affect the overall robustness of the system.

The incorporation of separation layer results in a geometry where the internal electric field is not fully compensated, i.e. the QW keeps a triangular potential profile. As a result, this 4-layer MQW system is more sensitive to changes in strain state versus the step-QW
The strain error bar in Fig. 2(c) illustrates the variation of the ISB transition when evolving from a structure fully strained on GaN to a structure fully strained on Al\textsubscript{0.1}Ga\textsubscript{0.9}N [19]. These error bars are comparable to those generated by uncertainties in the structural parameters. However, the MQWs are expected to evolve towards a minimum elastic energy configuration independently of the substrate [20], so that the uncertainty in the strain state of the structure (neglecting the initial relaxation) is much smaller (<± 0.025% variation of the in-plane lattice parameter) than the values simulated in Fig. 2(c) (± 0.12%).

An additional effect of the internal electric field is the localization of the wave functions, which affects the oscillator strength of the ISB transitions. In the 4-layer MQW design, both e\textsubscript{1} and e\textsubscript{2} are shifted in the [0001] direction [see Fig. 2(a)], which translates into a large oscillator strength. In contrast, the e\textsubscript{1} of the step-QW design experiences a shift in the [000-1] direction while e\textsubscript{2} shifts in the [0001] direction [see Fig. 1(a)], which creates a mismatch between the first and second localized states and translates into lower oscillator strength. Simulations show that the oscillator strength of the e\textsubscript{1}-e\textsubscript{2} transition within the 4-layer MQW system is about three times larger than that of the step-QW configuration.

AlGaN/GaN 40-period MQW structures following the 4-layer MQW design in Fig. 2(b) were synthesized by PAMBE on GaN templates on float-zone Si (111) to evade problems of substrate transparency [7]. These templates incorporate a complex buffer layer to manage the thermal expansion and lattice mismatch between GaN and Si. To simplify the structural characterization, identical samples were grown simultaneously on 1-μm-thick (0001)-oriented AlN-on-sapphire templates. During the deposition, the flux of active nitrogen was fixed at 0.32 ML/s and the growth temperature was ~720°C as deduced from the Ga desorption time. The three Al concentrations were obtained using two PAMBE cells, where one flux was set to 0.03×0.32 ML/s and the second was set to 0.07×0.32 ML/s. The
third Al concentration (10%) is obtained from the summation of the two fluxes. All the layers were deposited under self-regulated Ga-rich conditions [17] without growth interruptions. This growth method allows for planar growth of (Al)GaN heterostructures with interface sharpness on the atomic scale. The GaN layers were Si-doped, in order to populate the first electronic levels in the structure. Four samples with different doping concentrations (non-intentionally doped, and [Si] = 1.5×10^{19} \text{ cm}^{-3}, 3.0×10^{19} \text{ cm}^{-3}, \text{ and } 1.3×10^{20} \text{ cm}^{-3}) were synthesized. Figure 3 depicts four periods of the non-intentionally-doped structure viewed by high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) performed in an FEI Titan 80-300 microscope working at 200 kV. An Average Background Subtraction filter was used to discern the small variations in contrast between the layers, which are associated to the alloy compositional changes. The layer thicknesses obtained from the image are 42 ML Al_{0.03}Ga_{0.97}N/ 7 ML Al_{0.07}Ga_{0.93}N/ 7 ML GaN/ 14 ML Al_{0.1}Ga_{0.9}N.

High-resolution x-ray diffraction (HRXRD) measurements were carried out in a Seifert XRD 3003 PTS-HR system, with a beam concentrator in front of the Ge(220) four-bounce monochromator, and a Ge(220) two-bounce analyzer inserted in front of the detector. Figure 4 shows the $\omega$–$2\theta$ scan of the (0002) and (0004) x-ray reflections, and a reciprocal space map around the (-1015) reflection of the MQW structure doped with [Si] = 3.0×10^{19} \text{ cm}^{-3} and grown on an AlN-on-sapphire template. From these measurements, we extract a superlattice period of 19.7±0.2 nm. The experimental measurements in Figs. 4(a) and (b) are compared with a simulation using the X’Pert software, assuming an Al$_x$Ga$_{1-x}$N/ Al$_y$Ga$_{1-y}$N/ GaN/ Al$_{x+y}$Ga$_{1-x-y}$N (11.7 nm / 2 nm / 2 nm / 4 nm) MQW period, where the layer thickness are proportional to the number of monolayers measured by HAADF-STEM. From the reciprocal space map in Fig. 4(c) we extracted the relaxation state of the GaN buffer (86% relaxed on the AlN substrate), and the in-plane lattice parameter of the MQWs which is almost totally strained on the GaN buffer layer. By adjusting the Al mole fractions in the MQWs to get a
best fit to the (0002) and (0004) experimental diffractograms, we obtain $x = 0.025 \pm 0.005$ and $y = 0.07 \pm 0.01$. These values are in good agreement with the nominal values within the uncertainties considered above.

ISB absorption was probed by Fourier Transform Infrared spectroscopy (FTIR) with a Bruker V70v spectrometer using a Hg lamp and a Si bolometer. To account for the ISB transition selection rules, the sample facets were polished at a 60° angle to form a multi-pass waveguide with 3-4 total internal reflections. The four samples were tested in transmission mode using a far-IR polarizer to discern between TE and TM polarized light. Figure 5 shows the far-IR transmission spectra of the doped samples at low temperature ($T = 5-10$ K). In the sample with a lower doping level ($[\text{Si}] = 1.5 \times 10^{19}$ cm$^{-3}$), we observe a TM-polarized absorption dip centred around 27-29 µm ($\sim 14$ THz), which gets deeper and broader with increasing doping level. This absorption line is attributed to the transition from the first to the second electronic level in the QW, in good agreement with our theoretical calculations, which predict a transition at 26.5 µm. The normalized absorption line width for the sample with a doping level $[\text{Si}] = 1.5 \times 10^{19}$ cm$^{-3}$ is $\Delta f/f \sim 0.25$, which is a significant improvement in comparison to results in step QWs ($\Delta f/f \sim 0.5$ in ref. 14).

In conclusion, we have introduced a modified design of nitride-based ISB absorber for the THz spectral range consisting of a 4-layer MQW structure. Particular attention was paid to the robustness of the design with regarding the uncertainties associated to the growth. The structure has been realized by PAMBE, and shows distinct absorption of TM-polarized light centred around 27-30 µm ($\sim 14$ THz). This absorption gets deeper and broader with increasing doping levels, and is consistent with the predicted electronic transition between the first and the second electronic levels in the QWs.

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References


[19] The calculations also show that by decreasing the in-plane lattice parameter of the MQWs (compressive strain), a decrease in ISB energy is seen in the 4-layer design, which is consistent with a reduction of the internal electric field due to the smaller piezoelectric constants of GaN in comparison to AlN. In contrast, an increase of the ISB energy is seen in the step-QW design, due to the tendency of the first electronic level to keep confined close to the GaN layer.

Figure Captions

Figure 1. (a) Conduction band profile, and first ($e_1$) and second ($e_2$) electronic levels with their associated wave functions for an Al$_{0.05}$Ga$_{0.95}$N/Al$_{0.1}$Ga$_{0.9}$N/GaN (10 nm / 3 nm / 3 nm) step QW. (b) Shift of the wavefunction of the first electronic level associated to a variation of the Al concentration in the well layer. A higher step-well Al concentration (top) creates a more triangular well and increases confinement towards the GaN layer. A lower step-well Al concentration (down) creates a secondary confinement area at the opposite side of the well. To illustrate the effect clearly, the band profiles corresponds to Al$_{0.07}$Ga$_{0.93}$N/Al$_{0.1}$Ga$_{0.9}$N/GaN (10 nm / 3 nm / 3 nm) (top) and Al$_{0.03}$Ga$_{0.97}$N/Al$_{0.1}$Ga$_{0.9}$N/GaN (10 nm / 3 nm / 3 nm) (bottom) (c) Illustration of the robustness of the step-QW system. The dashed line indicates the nominal transition wavelength for an Al$_{0.05}$Ga$_{0.95}$N/Al$_{0.1}$Ga$_{0.9}$N/GaN (10 nm / 3 nm / 3 nm) step QW. The error bars represent the minimum and maximum values attributed to the uncertainties associated to growth. The barrier and GaN well thicknesses were changed from 3 nm to 2.5 nm and 3.5 nm. The barrier Al content was changed from 10% to 11% and 9%. The step well alloy was changed from 5% to 4.5% and 5.5% (±10%). The step well thickness was changed from 10 nm to 12 nm and 8 nm. The strain error bar illustrates the variation of the ISB transition when evolving from a structure fully strained on GaN to a structure fully strained on Al$_{0.1}$Ga$_{0.9}$N.

Figure 2. (a) Conduction band profile, and first ($e_1$) and second ($e_2$) electronic levels with their associated wave functions for an Al$_{0.1}$Ga$_{0.9}$N/GaN/Al$_{0.07}$Ga$_{0.93}$N/Al$_{0.03}$Ga$_{0.97}$N (4 nm / 2 nm / 2 nm / 12 nm) 4-layer-QW design. (b) Schematic
The dashed lines indicate the nominal transition wavelengths for an
$\text{Al}_{0.05}\text{Ga}_{0.95}\text{N}/\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}/\text{GaN}$ (10 nm / 3 nm / 3 nm) step QW and an
$\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}/\text{GaN}/\text{Al}_{0.07}\text{Ga}_{0.93}\text{N}/\text{Al}_{0.03}\text{Ga}_{0.97}\text{N}$ (4 nm / 2 nm / 2 nm / 12 nm) 4-
layer QW. The error bar represents the minimum and maximum values attributed
to the uncertainties associated to growth. In the 4-layer-QW, the barrier thickness
was changed from 2 nm to 1.5 nm and 2.5 nm. The barrier Al content was
changed from 10% to 11% and 9%. The GaN layer thickness was changed from
2 nm to 1.5 nm and 2.5 nm. The well thickness was changed from 12 nm to
11.5 nm and 12.5 nm, and its Al content was changed from 3% to 2.7% and 3.3%
(\pm 10\%). The separation layer thickness was changed from 2 nm to 1.5 nm and
2.5 nm, and its Al content was changed from 7% to 6.3% and 7.7%. The strain
error bar illustrates the variation of the ISB transition when evolving from a
structure fully strained on GaN to a structure fully strained on $\text{Al}_{0.1}\text{Ga}_{0.9}\text{N}$.

**Figure 3.** HAADF-STEM image showing several 4-layer-QWs in a non-intentionally
doped sample with structure described in Fig. 2(b).

**Figure 4.** HRXRD $\omega$–2$\theta$ scans of the (a) (0002) reflection and (b) (0004) reflection of the
MQW structure doped with $[\text{Si}] = 3.0 \times 10^{19}$ cm$^{-3}$ and grown on an AlN-on-
sapphire template, showing periodicity of 19.7±0.2 nm. The experimental
measurement is compared with simulations using the X’Pert Epitaxy software.
(c) Reciprocal space map around the (-1015) reflection of the MQW structure.

**Figure 5.** Far-IR transmission measurement of 4-layer MQWs with different doping levels
for TE- and TM-polarized light. The spectra have been normalized by the
response of a similar undoped superlattice which exhibits no ISB activity. The noise observed for wavelengths < 10 µm is due to the GaN Restrahlen absorption. The dip in TM-polarized transmission at 27-30 µm is assigned to the transition between the first and the second electronic levels in the QWs.