Nonpolar \textit{m}-plane GaN/AlGaN heterostructures with intersubband transitions in the 5–10 THz band

This content has been downloaded from IOPscience. Please scroll down to see the full text.
2015 Nanotechnology 26 435201
(http://iopscience.iop.org/0957-4484/26/43/435201)

View the table of contents for this issue, or go to the journal homepage for more

Download details:

IP Address: 129.11.21.2
This content was downloaded on 08/01/2016 at 21:46

Please note that terms and conditions apply.
Nonpolar $m$-plane GaN/AlGaN heterostructures with intersubband transitions in the 5–10THz band

C B Lim$^{1,2}$, A Ajay$^{1,2}$, C Bougerol$^{1,3}$, B Haas$^{1,2}$, J Schörmann$^4$, M Beeler$^{1,2}$, J Lähnemann$^{1,2}$, M Eickhoff$^4$ and E Monroy$^{1,2}$

$^1$ University Grenoble-Alpes, F-38000 Grenoble, France
$^2$ CEA, INAC-SP2M, 17 av. des Martyrs, F-38000 Grenoble, France
$^3$ CNRS, Institut Néel, 25 av. des Martyrs, F-38000 Grenoble, France
$^4$ I. Physikalisches Institut, Justus-Liebig-Universität Gießen, Heinrich-Buff-Ring 16, D-35392 Gießen, Germany

E-mail: caroline.lim@cea.fr

Received 29 June 2015, revised 1 September 2015
Accepted for publication 3 September 2015
Published 5 October 2015

Abstract

This paper assesses intersubband (ISB) transitions in the 1–10 THz frequency range in nonpolar $m$-plane GaN/AlGaN multi-quantum-wells deposited on free-standing semi-insulating GaN substrates. The quantum wells (QWs) were designed to contain two confined electronic levels, decoupled from the neighboring wells. Structural analysis reveals flat and regular QWs in the two perpendicular in-plane directions, with high-angle annular dark-field scanning transmission electron microscopy images showing inhomogeneities of the Al composition in the barriers along the growth axis. We do not observe extended structural defects (stacking faults or dislocations) introduced by the epitaxial process. Low-temperature ISB absorption from 1.5 to 9 THz (6.3–37.4 meV) is demonstrated, covering most of the 7–10 THz band forbidden to GaAs-based technologies.

Keywords: GaN, nonpolar, intersubband, THz, AlGaN, polarization

(Some figures may appear in colour only in the online journal)

1. Introduction

The development of compact solid-state THz sources is driven by its numerous applications in biological and medical sciences, industrial and pharmaceutical quality control, security screening and communication. GaAs-based quantum cascade lasers (QCLs) have proven their potential as emitters in the 0.85–5 THz range [1, 2], although the low maximum operation temperature remains a hurdle for their commercialization. There are two major processes that cause the degradation of population inversion in THz QCLs at higher temperatures [3]:

(i) Backfilling of the lower radiative state with electrons from the heavily populated injector, which occurs either by thermal excitation, or by reabsorption of non-equilibrium longitudinal optical (LO) phonons.

(ii) Thermally activated LO-phonon scattering, as electrons in the upper radiative state acquire sufficient in-plane kinetic energy to emit an LO-phonon and relax nonradiatively to the lower radiative state.

Both of these mechanisms sensitively depend on the electron gas temperature, which is 50–100 K higher than the lattice temperature during device operation. Thus, the GaAs LO phonon energy of 36 meV is a relatively low barrier for these thermally-activated phenomena. GaN, with an LO phonon energy of 92 meV (22.2 THz), opens prospects for room temperature THz lasers [4, 5]. Furthermore, phonon absorption in GaAs hinders the extension of these technologies towards the 5–10 THz range, and the blue shift of the Reststrahlen band in GaN opens the possibility of other intersubband (ISB) devices covering this 5–10 THz GaAs ‘forbidden’ band.
Table 1. Structural and optical characteristics of the $m$-plane GaN/AlGaN MQWs: QW and barrier thickness ($t_{QW}$ and $t_B$, respectively); Al composition of the barriers ($x_B$); MQW period measured by HR-XRD; FWHM of the $\omega$-scan of the $\overline{3300}$ x-ray reflection of the MQWs and of the GaN substrate, measured in the $c$ and $a$ directions ($\Delta \omega_c$ and $\Delta \omega_a$, respectively); simulated ISB transition energy; measured ISB transition energy window and central energy.

| Sample | $t_{QW}$ (nm) | $t_B$ (nm) | $x_B$ (%) | Period (nm) | $\omega$-scan FWHM MQW (arcsec) | $\omega$-scan FWHM GaN (arcsec) | Simulated ISB transition energy (meV) | Measured ISB transition energy window (meV) | Measured ISB transition central energy (meV) |
|--------|--------------|------------|----------|-------------|-------------------------------|-------------------------------|---------------------------------|---------------------------------|---------------------------------|
| S1     | 9.5          | 21.7       | 8        | 31.2        | $\Delta \omega_c = 35 \quad \Delta \omega_a = 34$ | $\Delta \omega_c = 34 \quad \Delta \omega_a = 39$ | 33                              | [13.4–37.4]                      | 25.4                            |
| S2     | 10.0         | 18.5       | 7.5      | 28.5        | $\Delta \omega_c = 38 \quad \Delta \omega_a = 35$ | $\Delta \omega_c = 35 \quad \Delta \omega_a = 36$ | 30.5                            | [18.1–36.1]                      | 27.1                            |
| S3     | 10.4         | 21.2       | 7.5      | 31.6        | $\Delta \omega_c = 22 \quad \Delta \omega_a = 31$ | $\Delta \omega_c = 24 \quad \Delta \omega_a = 39$ | 30.3                            | [6.3–36.3]                       | 21.3                            |
| S4     | 12.9         | 21.1       | 6        | 34.1        | $\Delta \omega_c = 38 \quad \Delta \omega_a = 28$ | $\Delta \omega_c = 42 \quad \Delta \omega_a = 29$ | 19.7                            | [7.9–33.9]                       | 20.9                            |
So far, studies on ISB transitions in group-III-nitride multi-quantum-wells (MQWs) have mostly focused on polar c-plane structures [6]. However, in this crystallographic orientation, optical properties are affected by a polarization-induced internal electric field, which renders ISB transition energies more sensitive to the strain state of the quantum wells (QWs) [7], and hampers the extension of ISB transitions towards far-infrared wavelengths [8]. Although the electric field can be partially compensated by the implementation of multi-layer QW designs [9–12], it is still a major hurdle for device design. The use of nonpolar crystallographic orientations leads to GaN/AlGaN heterostructures without internal electric field, which facilitates the device design while maintaining the benefits of GaN.

We have recently reported that the (1100) m plane is the most promising nonpolar crystallographic orientation for ISB applications, based on comparative results with the (1120) a plane [13]. Mid-infrared ISB absorption in the 4.0 to 5.8 µm (310 to 214 meV) range has been observed on m-plane GaN/AlGaN MQWs [13, 14] and photodetection at 7.5 and 9.3 µm (165 and 133 meV, respectively) has been demonstrated at 14 K using m-InGaN/(Al)GaN MQWs [15]. Recently, low-temperature (T = 9 K) ISB absorption has been shown in the 3.77–6.31 THz range (15.6–26.1 meV) using m-GaN/AlGaN MQWs [16]. However, ISB transitions in the 7–10 THz band, inaccessible to As-based technologies, have not been reported in nonpolar nitrdes yet. In this work, we investigate ISB transitions in nonpolar m-plane GaN/AlGaN MQWs in the 1–10 THz band.

2. Sample design and experimental section

A series of m-plane GaN/AlGaN MQW structures were designed using the Nextnano³ $8 \times 8$ k.p self-consistent Schrödinger–Poisson solver [17] with the material parameters described in [7]. A cell consisting of three QWs with periodic boundary conditions was used to model the MQW structure. Using this setup, the electronic decoupling of the QWs can be confirmed. The thickness of the barriers was fixed at approximately 20 nm to avoid coupling between QWs, and the QW widths were chosen to display ISB absorption between the ground conduction band level ($e_1$) and the first excited electronic level ($e_2$) in the 4.8–8 THz (19.7–33 meV) range. Additionally, the Al content of the barriers was varied to keep the two lowermost electronic levels confined in the QW and the third electronic level close to the continuum. Table 1 summarizes the m-plane MQW architectures considered in this paper, and figure 1 shows the calculated band diagram corresponding to sample S2 in table 1.

Structures consisting of 40 periods of GaN/AlGaN MQWs were deposited on free-standing semi-insulating m-GaN platelets sliced from (0001)-oriented GaN boules synthesized by hydride vapor phase epitaxy (resistivity $>10^4$ Ω·cm, dislocation density $<5 \times 10^6$ cm$^{-2}$). The MQW structures were capped with a 50 nm AlGaN layer with the same Al content as used for the barriers. The samples were grown by plasma-assisted molecular-beam epitaxy (PAMBE) at a substrate temperature of 720 °C and with a nitrogen-limited growth rate of 0.5 ML s$^{-1}$ ($\approx$450 nm h$^{-1}$). Growth was performed under the optimum conditions for c-GaN, i.e. slightly Ga-rich conditions [7, 18, 19], which are known to be compatible with m-plane growth [13, 20]. The GaN wells were homogeneously doped with silicon at a concentration of $3 \times 10^{18}$ cm$^{-3}$.

The samples were analyzed by High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy (HAADF-STEM) and High-Resolution Transmission Electron Microscopy (HR-TEM) performed in an FEI Titan Ultimate microscope operated at 200 kV.

![Figure 1](image1.png)  
Figure 1. Conduction band diagram with the three first electronic levels and their squared wavefunctions of a QW in the center of the active region of sample S2.

![Figure 2](image2.png)  
Figure 2. Cross-section HAADF-STEM images of sample S2 viewed (a) along (0001), and (b) along (1120). Layers with dark and bright contrast correspond to the AlGaN barriers and GaN QWs, respectively.
High-resolution x-ray diffraction (HR-XRD) measurements were done using a PANalytical X’Pert PRO MRD system. Experimental measurements were compared with simulations using the X’Pert Epitaxy software from PANalytical.

Fourier transform infrared spectroscopy (FTIR) was performed at 5 K using a Bruker V70v spectrometer equipped with a mercury lamp, a Si beam splitter, and a helium-cooled Si bolometer. Two pieces of each sample, each with a width of 4 mm and a thickness of 320 μm, were polished at 45° to form multipass waveguides. The pieces were placed face-to-face on the cold finger of a liquid helium-cooled cryostat, and were compared to pieces of m-GaN substrates prepared in the same fashion. All of the samples were tested in transmission mode using a far-infrared polarizer to discern between the transverse-electric (TE) and transverse-magnetic (TM) polarized light.

3. Results and discussion

To evaluate their structural quality, the samples were analyzed by HAADF-STEM and HR-TEM. Figures 2(a) and (b) show cross-section HAADF-STEM images of sample S2 viewed along (1120) and intensity profile along (0001). (b) High-resolution HAADF-STEM image of the barrier/QW interface showing that the variations of contrast in the image are not associated to structural defects.

From the intensity profile extracted from figure 3(a), the alloy fluctuations in the barriers can reach ±30% of the average concentration, which has been further confirmed by energy-dispersive x-ray spectroscopy (not shown).

The periodicity of the MQWs was assessed by HR-XRD. Figure 4 presents ω-2θ scans along the (3300) reflection of samples S1 and S4. Simulations assume the quantum structures fully strained on the GaN substrate.
full width at half maximum of the rocking curves was measured for the substrate and the MQW zero-order (33̄000) reflection with $\phi = 0^\circ$ and $\phi = 90^\circ$ ($\Delta\omega_c$ and $\Delta\omega_a$, respectively). These broadening values provide information about the sample mosaicity in the $c$ and $a$ directions, respectively. Both $\Delta\omega_c$ and $\Delta\omega_a$ remain in the $30 \pm 8$ arcsec range for all the MQWs, and these values are similar to those measured for the substrate reflections. This result is consistent with the absence of epitaxially-generated defects in the transmission electron microscopy images.

To probe the ISB absorption of the MQWs, characterization by FTIR spectroscopy was performed at 5 K. Using sample S2 as an example, figure 5(a) illustrates typical TE- and TM-polarized THz transmission measurements, and figure 5(b) compares the same transmission spectrum for TM-polarized light with that of the substrate. The apparent noise superimposed on all the spectra is an oscillation with nearly-regular periodicity in energy (see magnified view in the inset of figure 5(a)), which is assigned to a Fabry–Pérot interference. Using the refractive index of GaN in the far-infrared range from [21], the cavity length associated to the interference is $\approx 350 \mu m$, corresponding to the overall thickness of the samples. The transmission spectra for TE-polarized light present additional Fabry–Pérot oscillations associated to the MQW layers. In contrast, the transmission spectrum for TM-polarized light exhibits a broad dip, in the $3$–$8$ THz range in the case of sample S2, which is assigned to ISB absorption, following the polarization selection rule.

Figure 6 presents the normalized absorbance of samples S1–S4 for TM-polarized light, extracted from the transmission measurements. TM-polarized absorption is observed over a broad spectral window of normalized bandwidth close to 1, whose extreme values and central energy are summarized in table 1. The central energy decreasing from $27.1$ to $20.9$ meV (6.5–5 THz) as the QW width increases is consistent with the trend of the simulations. The broad absorption bands are consistent with the doping density in the QWs of around $n_S = 3 \times 10^{12} \text{cm}^{-2}$, i.e. at least three times higher.
than in [16], as illustrated in the inset of figure 5(b). The samples show ISB absorption ranging from 6.3 to 37.4 meV (1.5–9 THz), providing experimental evidence that ISB transitions in GaN MQWs can cover the THz spectral range forbidden to GaAs.

4. Conclusions

In summary, we have designed a series of nonpolar m-plane GaN/AlGaN MQWs by varying the QW thicknesses and Al compositions to separate the two confined electronic levels by 20–33 meV (corresponding to 4.8–8 THz transitions), and decouple these transitions from the neighboring wells. The samples were grown by PAMBE on free-standing semi-insulating GaN substrates, and the structural analysis showed MQWs composed of flat and regular layers in the two perpendicular in-plane directions, with inhomogeneities of the Al composition in the barriers along the growth axis. Extended defects introduced by the epitaxial process, such as stacking faults or dislocations, were not observed. Optically, the structures display low-temperature ISB absorption with a normalized bandwidth close to 1, which is attributed to the high doping level. This absorption occurs in the 6.3–37.4 meV (1.5–9 THz) range, providing an experimental demonstration of the possibility for GaN to cover a large part of the 7–10 THz band forbidden to GaAs-based technologies.

Acknowledgments

Thanks are due to N Molland for sample preparation by focused ion beam at the NanoCharacterization Platform (PFNC) in CEA-Minatec Grenoble. The free-standing semi-insulating M-GaN substrates were kindly supplied by Suzhou Nanowin Science and Technology Co., Ltd (NANOWIN). This work is supported by the EU ERC-StG ‘TeraGaN’ (#278428) project. AA acknowledges financial support by the French National Research Agency via the GaNEX program (ANR-11-LABX-0014).

References

[1] Köhler R, Tredicucci A, Beltram F, Beere H E, Linfield E H, Davies A G, Ritchie D A, Iotti R C and Rossi F 2002 Terahertz semiconductor-heterostructure laser Nature 417 156–9
[2] Williams B S 2007 Terahertz quantum-cascade lasers Nat. Photonics 1 517–25
[3] Han Y J and Cao J C 2012 Temperature dependence of electron transport on a bound-to-continuum terahertz quantum cascade laser Semicond. Sci. Technol. 27 015002
[4] Bellotti E, Driscoll K, Moustakas T D and Puella R 2009 Monte carlo simulation of terahertz quantum cascade laser structures based on wide-bandgap semiconductors J. Appl. Phys. 105 113103
[5] Sun G, Khurgin J B and Tsai D P 2013 Spoof plasmon waveguide enabled ultrathin room temperature THz GaN quantum cascade laser: a feasibility study Opt. Express 21 28054
[6] Beeler M, Trichas E and Monroy E 2013 III-nitride semiconductors for intersubband optoelectronics: a review Semicond. Sci. Technol. 28 074022
[7] Kandaswamy P K et al 2008 GaN/AlN short-period superlattices for intersubband optoelectronics: a systematic study of their epitaxial growth, design, and performance J. Appl. Phys. 104 093501
[8] Kandaswamy P K, Machhadani H, Bougerol C, Sakr S, Tchernycheva M, Julien F H and Monroy E 2009 Mid-infrared intersubband absorption in GaN/AlGaN superlattices on Si(111) templates Appl. Phys. Lett. 95 141911
[9] Machhadani H, Kotsar Y, Sakr S, Tchernycheva M, Colombelli R, Mangeney J, Bellett-Amalric E, Sarigianidou M, Monroy E and Julien F H 2010 Terahertz inter-subband absorption in GaN/AlGaN step quantum wells Appl. Phys. Lett. 97 191101
[10] Sadradjat F F, Zhang W, Woodward J, Durmaz H, Moustakas T D and Puella R 2012 Far-infrared intersubband photodetectors based on double-step III-nitride quantum wells Appl. Phys. Lett. 100 241113
[11] Beeler M, Bougerol C, Bellett-Amalric E and Monroy E 2013 Terahertz absorbing AlGaN/GaN multi-quantum-wells: demonstration of a robust 4-layer design Appl. Phys. Lett. 103 091108
[12] Beeler M, Bougerol C, Bellett-Amalric E and Monroy E 2014 Pseudo-square AlGaN/GaN quantum wells for terahertz absorption Appl. Phys. Lett. 105 131106
[13] Lim C B, Beeler M, Ajay A, Lähnemann J, Bellett-Amalric E, Bougerol C and Monroy E 2015 Intersubband transitions in nonpolar GaN/Al(Ga)N heterostructures in the short and mid-wavelength infrared regions J. Appl. Phys. 118 014309
[14] Kotani T, Arita M and Arakawa Y 2014 Observation of mid-infrared intersubband absorption in non-polar m-plane AlGaN/GaN multiple quantum wells Appl. Phys. Lett. 105 261108
[15] Pesach A, Gross E, Huang C-Y, Lin Y-D, Vardi A, Schacham S E, Nakamura S and Bahir G 2013 Non-polar m-plane intersubband based InGaN/AlGaN quantum well infrared photodetectors Appl. Phys. Lett. 103 022110
[16] Edmonds C, Shao J, Shirazi-HD M, Manfra M J and Malis O 2014 Terahertz intersubband absorption in non-polar m-plane AlGaN/GaN quantum wells Appl. Phys. Lett. 105 201109
[17] Birner S, Zibold T, Andlauer T, Kubis T, Sabathil M, Trellakis A and Vogl P 2007 nextnano: general purpose 3D simulations IEEE Trans. Electron Devices 54 2137–42
[18] Heying B, Averbeck R, Chen L, F, Haus E, Riechert H and Speck J S 2000 Control of GaN surface morphologies using plasma-assisted molecular beam epitaxy J. Appl. Phys. 88 1855–60
[19] Adelmann C, Brault J, Mula G, Daudin B, Lymperakis L and Neugebauer J 2003 Gallium adsorption on (0001) GaN surfaces Phys. Rev. B 67 165419
[20] Shao J, Zakharov D N, Edmonds C, Malis O and Manfra M J 2013 Homogeneous AlGaN/GaN superlattices grown on free-standing (11’00) GaN substrates by plasma-assisted molecular beam epitaxy Appl. Phys. Lett. 103 232103
[21] Ibáñez J, Hernández S, Alarcón-Lladó E, Cuscó R, Artús L, Novikov S V, Foxon C T and Calleja E 2008 Far-infrared transmission in GaN, AlN, and AlGaN thin films grown by molecular beam epitaxy J. Appl. Phys. 104 033544