Bandgap, electrical and structural properties of thick InN (0001) films grown under optimal conditions

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Abstract. The improvement potential for the structural, electrical and optoelectronic properties of heteroepitaxial InN-on-GaN (0001) films by using optimal conditions (substrate temperature, In and N fluxes) of plasma-assisted molecular beam epitaxy and increasing the epilayer thickness to few micrometres has been investigated. The increase of InN thickness to 3.7 µm resulted to a-type component threading dislocation density of 6x10^9 cm^-2 and directly measured electron mobility of 2330 cm²/Vs and concentration of 4.5x10^17 cm^-3. The optical bandgap of this film at 300K was 0.637 eV. However, a degradation in the integrity of the interfacial InN/GaN region was observed in films thicker than 1 µm, with the formation of voids and the nucleation of microcracks, which may be related to strain relaxation or thermal decomposition.

1. Introduction

InN is predicted to exhibit supreme transport properties, such as electron low-field mobility of 14,000 cm²/Vs in undoped InN [1] and peak drift velocity of 5.3-5.6 x 10^7 cm³ [1, 2] for steady-state conditions. Even more promising for nanoelectronic devices are the transient transport characteristics of InN, with an overshoot electron drift velocity that may approach 10⁸ cm/s [2].

Recent works [3, 4] have demonstrated the feasibility of realizing InN channel transistors but also the negative effect of heteroepitaxial defects on the transport properties and the breakdown electric field in an InN channel. In addition, the accurate determination of InN bandgap [5] is complicated by not very abrupt absorption edge, with energy shift due to conduction band filling [6] by high electron concentration.

The present work explores the potential to control and reduce the above material problems that limit the realization of actual properties of InN crystal and their exploitation into state-of-the-art devices. The approach was to grow InN films under the optimal conditions for two-dimensional growth [7] without InN decomposition [8] and increase the epilayer thickness to reduce the threading dislocation (TD) density [9]. InN films with thickness 0.78-3.7 µm were grown and characterized. A quantitative evaluation of the InN TD density, electron concentration and mobility, and bandgap is presented for low defect density InN, as well as the dependence of electrical properties on TDs.
2. Experimental

Three InN films with thickness of 0.78, 1.45 and 3.7 μm have been grown by Plasma-Assisted Molecular Beam Epitaxy (PAMBE) on high resistivity 3 μm GaN-on-Al₂O₃ (0001) templates. InN growth was carried out under stoichiometric In/N flux ratio that favors the two-dimensional growth mechanism [7] and at the maximum substrate temperature (Tsub) allowing thermal stability of InN [8].

The FWHM values of High Resolution X-ray diffraction (HRXRD) ω-scans for symmetric (0002) and skew symmetric (11-24) diffractions were used to estimate the density of TDs with c-type (screw) and a-type (edge) component, respectively, using the formulas of [10, 11]. HRXRD measurements utilizing the extended Bond method were used to determine the a and c lattice constants. Carrier concentration and mobility were determined by Hall-effect (at magnetic field of 0.32T) and conductivity measurements, in a Van der Pauw configuration, using 4 indium dot contacts. The bandgap and optoelectronic properties were studied by photoluminescence spectroscopy (PL) at 15K on all samples and optical transmittance spectroscopy (T) at 300K on the thickest film. Scanning Electron Microscopy (SEM) was used for inspection of the samples’ cross-section and thickness measurements.

3. Results and discussion

Table 1 presents the HRXRD results for the three samples, as well as the estimated densities of TDs [10, 11]. The FWHM values of (0002) ω-scans, which are indicative of crystal twist and the density of TDs with c-type component, were similar and did not exhibit any InN thickness dependence. However, a clear FWHM reduction with epilayer thickness was observed in ω-scans for the inclined (11-24) planes, which are broadened also by crystal twist and the density of a-type component TDs. The calculated a-type TD density dropped to 6x10⁷ cm⁻² in the 3.7 μm InN film. The lattice constants for all samples were similar and close to the strain-free state [12].

Table 1: Sample thickness, HRXRD FWHMs and calculated TD densities for the three InN samples. The a and c lattice constants are also shown.

| Sample | Thickness (μm) | XRD FWHM (arcsec) | TD density (cm⁻²) | c (Å) | a (Å) |
|--------|----------------|--------------------|-------------------|-------|-------|
|        | (0002)         | (11-24)            | c-component       | a-component |
| A      | 0.78           | 415                | 2.8 x 10⁸         | 2.6 x 10¹⁰  | 5.705 | 3.533 |
| B      | 1.45           | 360                | 2.1 x 10⁸         | 1.5 x 10¹⁰  | 5.701 | 3.536 |
| C      | 3.7            | 425                | 2.9 x 10⁸         | 6.0 x 10⁸    | 5.702 | 3.533 |

The directly measured electron density (N_Smeas) and mobility (μ_Smeas) values, by Hall effect experiments, are shown in Table 2. These values include the contribution of electrons from the bulk of InN film and surface/interfacial electron accumulation [13]. The measurements were analysed according to formulas of ref. [14], assuming a total electron accumulation density N_Sref = 2.8 x 10¹³ cm⁻² (estimated by extrapolating results to zero thickness) with mobility μ ≈ 320 cm²/Vs according to [13]. The extracted electron concentration (N_D) and mobility (μ_bulk) for the bulk of the InN film are given in Table 2.

Table 2: Directly measured electron densities (N_Smeas) and mobilities (μ_Smeas) at 300K, by Hall effect experiments and extracted electron concentration (N_D) and mobility (μ_bulk) for the bulk of the InN film.

| Sample | Measured values | Extracted bulk values |
|--------|-----------------|-----------------------|
|        | N_Smeas (cm⁻³) | μ_Smeas (cm²/Vs) | N_D (cm⁻³) | μ_bulk (cm²/Vs) |
| A      | 8.1 x 10¹⁷      | 1939                 | 7.0 x 10¹⁷ | 2067             |
| B      | 5.2 x 10¹⁷      | 2164                 | 4.7 x 10¹⁷ | 2271             |
| C      | 4.5 x 10¹⁷      | 2332                 | 4.3 x 10¹⁷ | 2379             |
The correlation of the electrical and structural results supports a qualitative conclusion for reduction of $N_D$ and increase of $\mu_{\text{bulk}}$ with reducing the density of a-type component TDs by increasing the InN epilayer thickness, as shown in Figure 1. However, these results do not fit in a quantitative physical model assuming dominant role of TDs in electron scattering and contribution of donors, suggesting complex influences on the properties of InN films and possible growth variations.

The 15K PL spectra of the three samples were in qualitative agreement with the determined differences in $N_D$. The optical band gap ($E_{\text{opt}}$) was estimated from transmittance (T) measurements on the thickest (3.7 $\mu$m) film, using the absorption $\alpha(E)$ approximation [5]

$$\alpha(E) = \frac{\alpha_0}{1 + \exp(E_{\text{opt}} - E)/\Delta}$$

(1)

where $\alpha(E)$ is derived from $T$ through the relation $\alpha \approx \frac{1}{d} \ln(T)$ where $\Delta$ is the Urbach broadening factor. Figure 2 shows the experimental $\alpha(E)$ and its fitting using Eq. (1). The estimated $E_{\text{opt}}$ is 0.637 eV with 20 meV broadening. Considering a Burstein-Moss energy shift [6,15] of 14 meV for 4.3 x $10^{17}$ cm$^{-3}$ electrons, the calculated fundamental energy gap at 300K is 0.623 eV, in good agreement with some literature results [11,16]. According to the temperature effect on the bandgap [5,15], the fundamental bandgap at 15K will be 0.672 eV, which is in reasonable agreement with the 15K PL spectrum of this sample (inset of Figure 2). Possible non-uniformity of the carrier concentration or strain, along the growth axis or laterally, may explain any small deviations observed.

The formation of voids and microcracks at or near the InN/GaN interface, as shown in Figure 3, may explain any small deviations observed.
The thicker than 1 μm InN films exhibited degradation in the integrity of the interfacial InN/GaN region. Figure 3 shows cross sectional SEM micrographs of sample B, exhibiting voids at the InN/GaN interface and microcracks parallel to the (0001) surface plane within a region of 100 nm above the interface. These phenomena may be related to strain relaxation mechanisms and/or thermal decomposition due to the marginally high growth temperature.

4. Conclusions
A 3.7 μm InN film, grown on GaN (0001) under the optimal PAMBE conditions, exhibited a significant structural improvement as evident by a FWHM of 836 arcsec for the HRXRD skew symmetric (11-24) ω-scan, which leads to an estimated density of 6 x 10^9 cm^-2 for a-type TDs.

This lead to reduction of the directly measured electron concentration to 4.5 x 10^17 cm^-3 with increased mobility to 2330 cm²/Vs. The optical bandgap of this film at 300K was 0.637 eV, which corresponds to InN fundamental bandgap of 0.623 eV.

A full strain relaxation was observed in all films. However, this might be also related to a degradation of the integrity of the interfacial region observed by SEM on films thicker than 1 μm.

Acknowledgements
This work has been supported partially by the European Research Infrastructure NFFA-Europe, funded by EU’s H2020 framework program (grant n. 654360) and the Hellenic INNOVATION-EL infrastructure (MIS 5002772).

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