

Atom Probe Tomography Quantification of Alloy Fluctuations in (Al,In,Ga)N

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As compared with ternary nitrides systems such as InGaN, AlGaN and InAlN, quaternary (Al,In,Ga)N benefit from a significant increase in design freedom. Indeed changes in the III site atomic fraction allows for an independent choice of either band-gap, lattice constant or polarization which is particularly interesting in the field of solid state lighting, radio frequency (RF), and power electronics. Controlling the homogeneity of the alloy is of great interest to understand the physical properties of (Al,In,Ga)N based devices [1,2]. Atom probe tomography (APT) has demonstrated its ability to evidence alloy fluctuations in ternary nitride [3]. However, detection artifacts in APT could lead to misinterpretation of the actual compositions [4,5]. Calibration experiments are required to find evaporation parameters which enable for an accurate quantification of all III site atoms in (Al,In,Ga)N.

Two samples with different aluminum, indium and gallium III site fractions and grown by metalorganic chemical vapour deposition (MOCVD) are investigated. The MOCVD growth reactor is a Veeco P75 Turbodisc reactor and the samples were grown on Ga-face GaN on sapphire substrates. APT tips were prepared by dual-beam FIB/SEM by the standard lift-out method and APT analysis were performed with a Cameca 3000X HR Local Electrode Atom Probe (LEAP) operated in laser-pulse mode. SIMS profiles were obtained on a Physical Electronics 6650 Dynamic Secondary Ion Mass Spectrometry (SIMS) tool to get concentrations references for APT measurements.

A reference sample, $\text{Al}_{0.260}\text{Ga}_{0.668}\text{In}_{0.072}$, was quantified by Rutherford Backscattering Spectroscopy and then measured via SIMS. The relative signal intensities were tuned to yield identical results between measurement techniques. The compositions of the samples in this study were determined with this same calibration. The average values are $\text{Al}_{0.35}\text{In}_{0.14}\text{Ga}_{0.51}\text{N}$ and $\text{Al}_{0.36}\text{In}_{0.10}\text{Ga}_{0.54}\text{N}$ as shown as dotted lines on figure 1. The evolution of the III site atomic ratios measured by APT with different evaporation conditions is displayed on figure 1 for the two samples. The $\text{Ga}^{++}/\text{Ga}^+$ ratio is used as an indicator of the intensity of the field applied on the APT tips. As shown on the figure, the intensity of the field does not have an influence on the measured III site ratios. However, as a comparison to SIMS, APT seems to detect more In and Ga to the detriment of Al. In ternary AlGaN, Ga is always under-estimated at high fields and further samples will be investigated to explain why the reverse is observed in this study.

A 3D reconstruction of GaN/InAlGaN/GaN is presented in figure 2(a) where all III site atoms can be observed. Figure 2(b) show the Ga, Al and In fractions of the bins for the $40 \times 40 \times 40 \text{ nm}^3$ volume shown on figure 2(a). The volume is subdivided in sampling volumes of 200 atoms in which the III site fractions are measured. The experimental distribution are compared to the binomial distribution expected for random alloys thanks to the χ^2 test. p values of 0.85 for Ga, 0.31 for Al and 0.74 for In confirm that all III site atoms are randomly distributed.

Quantification of (Al,In,Ga)N by APT remains a challenge due to the incomplete understanding of field evaporation of semiconductors composed of such a high number of different atoms. Still, direct observation of a binomial alloy distribution confirms the results from reference [1], which had assumed a Gaussian distribution in Schottky barrier heights for the sample [7].

References:

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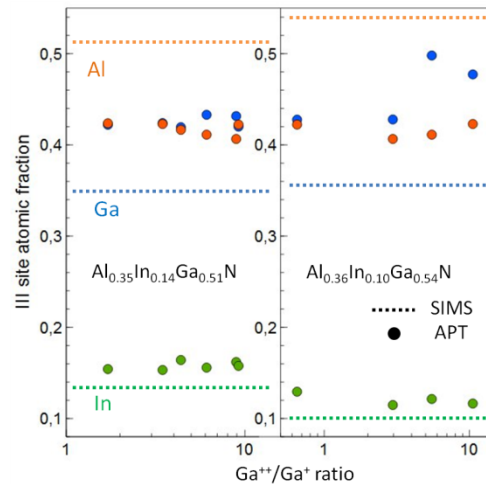


Figure 1. (a) Evolution of the III site atomic ratios of Al, In and Ga in $\text{Al}_{0.35}\text{In}_{0.14}\text{Ga}_{0.51}\text{N}$ and $\text{Al}_{0.36}\text{In}_{0.10}\text{Ga}_{0.54}\text{N}$ with respect to the intensity of the field applied on the specimens. SIMS values are displayed as dotted lines for the two samples.

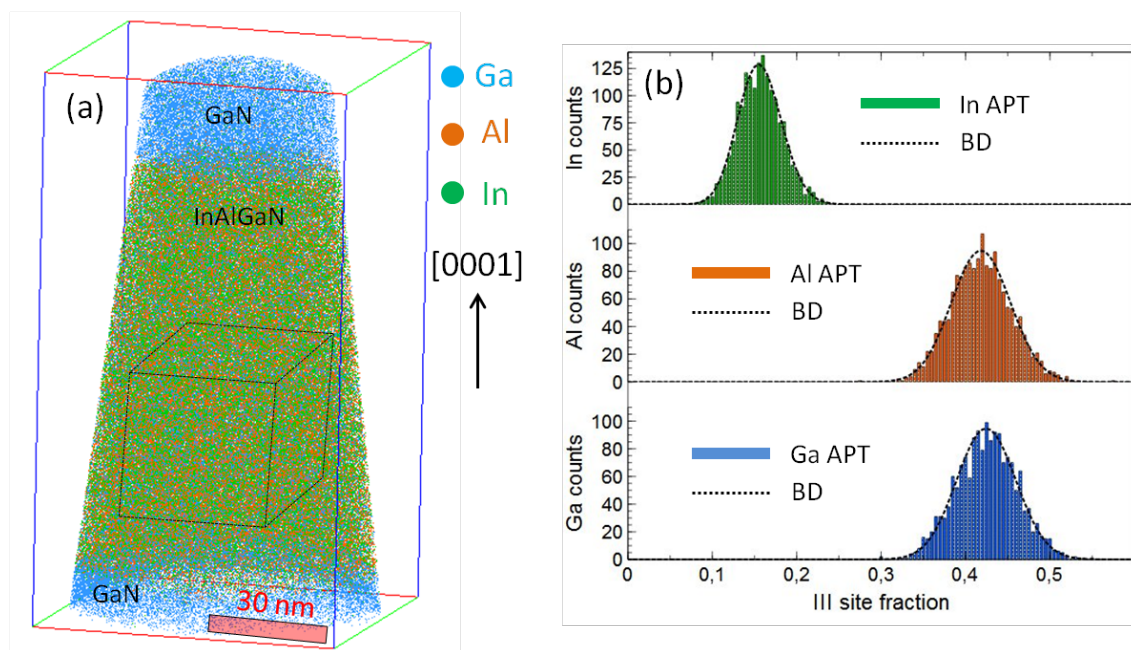


Figure 2. (a) APT 3D reconstruction of $\text{Al}_{0.35}\text{In}_{0.14}\text{Ga}_{0.51}\text{N}$ ($\text{Al}_{0.42}\text{In}_{0.16}\text{Ga}_{0.42}\text{N}$ according to APT). (b) Gallium, Aluminum and Indium fraction frequency distributions (histograms) obtained in the sampling volume in (a) showing good agreement with the binomial distribution (dotted curves).