Extended Abstracts of the 2019 International Conference on Solid State Devices and Materials, Nagoya, 2019, pp263-264

Investigation of doping in III-nitrides by combining atom probe tomography and EDX spectroscopy

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Abstract

Atom Probe Tomography (APT) has emerged as a reliable chemical analysis technique as it provides access to the chemical composition of the analyzed material together with the 3D distribution of the atoms in a tip-shaped specimen with hundred-nanometer diameter. Moreover, combining APT with EDX enables first to overcome some artefacts inherent to APT to get quantitative data but also permits to investigate nanostructures from nano- to micro-scale. We have successfully applied these techniques for studying both n-type (Ge) and p-type (Mg) doping of AlGaN layers.

1. Introduction

Further enhancement of the performance of advanced nitride-based optoelectronic devices requires control of the material morphology as well as fine tuning of composition and doping. As the electrical conductivity depends on the type, quantity, and distribution of dopants, understanding the behavior of dopants requires the determination of their concentration and 3D spatial distribution at the nano-scale. This represents a real challenge which can only be addressed by combining several characterization techniques.

2. Experimental methods

Atom Probe Tomography (APT) appears as the technique of choice as it provides the 3D distribution of atomic species at the nanometer-scale in a tip-shaped specimen. However, in the case of nitrides, some composition biases linked to the experimental parameters (temperature, laser wavelength and power, evaporation field) have been reported [1-2] which makes the quantitative dopant concentration difficult to obtain. In that context, combining APT with EDX spectroscopy enables to overcome these artefacts and grants access to both nano- and micro-scale chemical analysis [3]. To quantify low level concentrations by EDX, the major challenge is to extract a low intensity signal from a relatively high background. We have recently overcome this issue by using a dedicated set-up for data recording, and by developing a new analytical procedure based on spectrum normalization to an undoped reference [4].

3. Results

Recently, we have successfully applied APT and EDX for studying n-type (Ge) and p-type (Mg) (Al)GaN layers grown by MBE and MOCVD, respectively.

n-type doping: Ge

Although Si has long been the preferred n-type dopant for GaN, Ge is currently under consideration, particularly for applications requiring dopant concentrations around or higher than 10^{19} cm^{-3}. In GaN, from both APT and EDX experiments, we have observed a homogeneous distribution of Ge atoms up to concentrations of 10^{20} cm^{-3} [5]. In the case of $Al_xGa_{1-x}N$ layers, we found that even at these high doping levels Ge does not induce any structural degradation in layers with $x < 0.15$. However, for higher Al compositions, clustering of Ge forming crystallites was observed from EDX maps (fig.1). This might explain the gradual decrease of the carrier concentration when increasing the Al mole fraction above $x = 0.24$ revealed from Hall measurements [6].

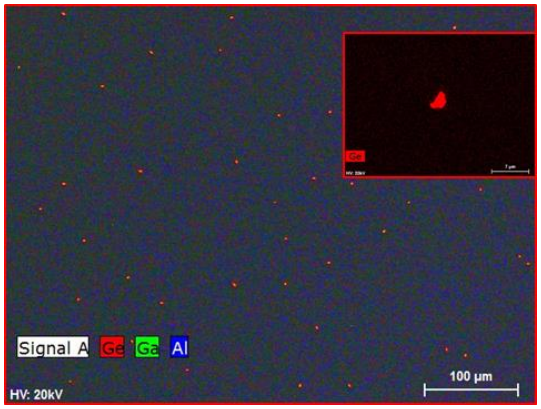

Fig.1 EDX map of a sample of nominal composition $Al_{.36}Ga_{.64}N$ Ge crystallites appear in red (zoom given in inset)

p-type doping: Mg

Mg is the only efficient p-type dopant though it has a relatively high ionization energy. Thus a concentration in the range 10^{19} to 10^{20} cm^{-3} is needed to get a hole concentration above 10^{17} cm^{-3} at room temperature. However, for metalorganic chemical vapor deposition growth (MOVCD), a decrease in the carrier concentration has been reported by several authors for nominal Mg concentrations above ~3 x10^{19} at.cm^{-3} [7]. This has been tentatively assigned to defects such as Mg-V_N complexes, acting as compensation centers [8], and/or to Mg-rich pyramidal inversion domains (PIDs) which are formed for these concentrations [9]. We have analyzed a GaN stack with different Mg concentrations (Fig.2 left). APT reconstruction (Fig.2 middle and right) shows Mg rich clusters for the two doped layers which have been evaporated. These are very likely related to the PIDs even though the spatial resolution is not high enough to evidence the expected triangular shape of the PIDs. Using the maximum separation distance technique based on the calculation of the distance between the first nearest neighbors of solute atoms, a cluster identification has been performed. We have observed that the cluster density increases with the dopant concentration. Moreover, it has been possible to access to the dopant concentration in the clusters as well as in the matrix around them. From these data, the concentration of dopants which could potentially participate to the electrical activity has been estimated [10].

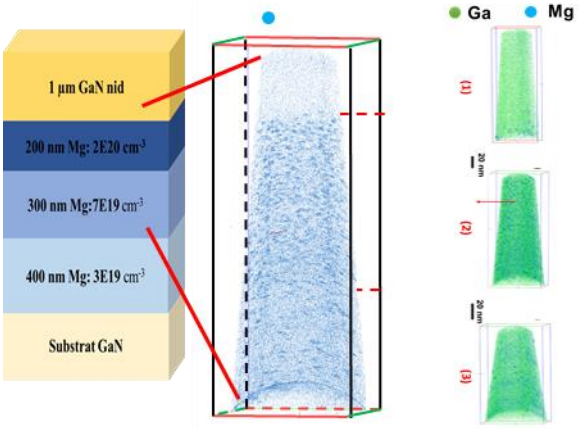

Fig. 2 Left: Schematic of the sample investigated by APT. Middle: APT reconstruction from a zone of the sample identified by the red lines. Right: reconstructed volumes in the layers with different doping, showing the presence of Mg-rich clusters in the doped-layers

3. Conclusions

Both EDX and APT can detect dopants for concentrations above ≅ 10^{19} at.cm^{-3} but quantification requires accurate calibrations and perfect control of the experimental conditions. APT is a very powerful technique to detect and analyze the 3D distribution of dopants in the case of inhomogeneous distributions, even at nm scale, giving access to the composition of the different parts of the materials. However in the case of nitrides, evaporation is challenging and strongly depends on the experimental conditions. Therefore concentration of all elements cannot always be obtained**.** EDX enables an investigation at different spatial scales and is usually easier and faster to perform with no need of sophisticated sample preparation. Combining both techniques is a very efficient way to reach a quantitative analysis of the spatial distribution of dopants in 3D at the nanoscale.

Acknowledgements

L. Amichi would like to thank the GaNex organization (ANR-11-LABX-0014) for funding her Phd position. GaNex belongs to the public funded "Investissements d'avenir" program managed by the French ANR Agency

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