

## Assessing the Composition of Wide Bandgap Compound Semiconductors by Atom Probe Tomography: A Metrological Problem.

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With the increasing application of Atom Probe Tomography (APT) to the study of nanoscale semiconductor systems for electronics, optoelectronics and related domains, experimentalists face the problem of measuring elemental compositions with high accuracy. This is possible under the hypothesis that all elemental species are field evaporated and detected with the same rate. However, recent studies have shown that for certain materials this hypothesis does not hold [1-6]. It becomes then necessary to explore the field evaporation behavior of compound semiconductors in order to (i) assess whether there is a bias in the compositional measurement (ii) understand what are the physical mechanisms inducing a bias and (iii) correct the bias, if possible. This contribution illustrates the results obtained on binary wide bandgap semiconductors, such as GaN, AlN, ZnO and MgO, and on selected ternary alloys, such as  $\text{Al}_y\text{Ga}_{1-y}\text{N}$  and  $\text{Mg}_y\text{Zn}_{1-y}\text{O}$ .

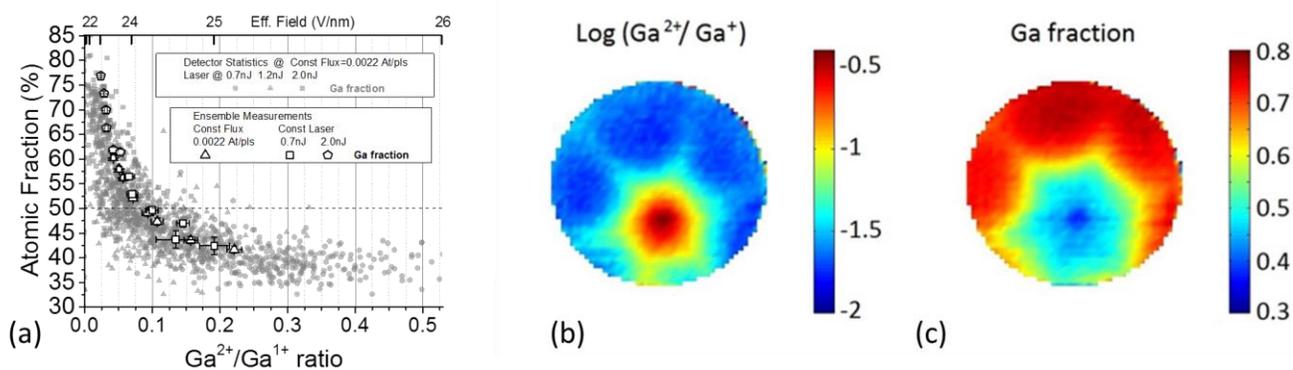
The study of the binary systems has been carried out by performing separate analyses either at constant detection rate or varying one experimental parameter at once (laser intensity and DC applied bias). The set of measurements shows that the main factor driving the compositional biases is the surface field, which can be estimated through the charge state ratios ( $\text{Me}^{2+}/\text{Me}^{1+}$ ) of the metallic constituent element Me according to the post-ionization theory [7]. In general, all measurements exhibit an excess of metallic element at low field, and – with the notable exception of MgO – an excess of N or O at high field (Fig. 1-(a)). This behavior can be partially explained through preferential evaporation of the metallic element at high field, while the mechanism responsible of the loss of N or O at low field is still under debate. Neutral evaporation or production of neutral N and O upon dissociation of more complex molecules have been proposed [1-6,8]. The field dependence of the measured composition also implies that the measurement is not uniform on the tip surface, as the field itself varies due to the specimen geometry and crystallography (Fig. 1 (b,c)).

The above mentioned binary systems have an *a priori* known composition, but this is not the case for ternary alloys and doped materials. In these technologically important cases, it becomes essential to know under which conditions APT may yield an accurate measurement of the composition. For ternary alloys such as  $\text{Al}_y\text{Ga}_{1-y}\text{N}$  and  $\text{Mg}_y\text{Zn}_{1-y}\text{O}$ , we studied the conditions under which the measurement of the

site fraction  $y$  is accurate. The study has been performed on GaN/  $\text{Al}_y\text{Ga}_{1-y}\text{N}$  and on ZnO/  $\text{Mg}_y\text{Zn}_{1-y}\text{O}$  quantum heterostructures. The surface field progressively decreased during the APT analyses at constant detection rate due to the increase of the surface imaged on the detector in sample tips with non-zero shank angle. Due to the different chemical properties of the metallic constituents of the alloys, Ga and Zn undergo preferential evaporation more easily than Al and Mg. As a result, the measured site fraction in the alloys is found to increase with the surface field. The accurate site fraction of these alloys may thus be determined performing the measurement at low field. This, however, could imply a degradation of spatial resolution. In order to circumvent this problem, a statistical correction scheme was developed, aiming to restore both the average value of the site fraction  $y$  and its standard deviation  $\sigma_y$ , in order to correctly reproduce the spatial behavior and the amplitude of the alloy fluctuations. The correction procedure has been tested for  $\text{Al}_{0.25}\text{Ga}_{0.75}\text{N}$  by photoluminescence (PL) spectroscopy. This technique allows indeed measuring the emission and localization energy of the photons emitted within the alloy, two quantities that depend on the average site fraction and on the alloy fluctuations [9]. [10]

#### References:

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**Figure 1.** Correlation of the measured Ga fraction in GaN with the charge state ratio  $\text{Ga}^{2+}/\text{Ga}^{1+}$ , related to the surface electric field. (a) Set of measurements acquired varying the experimental parameters (open black symbols) and results from the detector statistics for three different datasets acquired with constant experimental parameters (grey symbols). Distribution of (a) the  $\text{Ga}^{2+}/\text{Ga}^{1+}$  charge state ratio and (b) Ga fraction on the detector space.