Electron energy loss near edge structure (ELNES) of $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys

M-M Soumelidou$^1$, J. Kioseoglou$^1$, H. Kirmse$^2$, Th. Karakostas$^1$ and Ph. Komninou$^1$

1. Physics Department, Aristotle University of Thessaloniki, GR-54124 Thessaloniki, Greece
2. Institute of Physics, Humboldt University of Berlin, D-12489 Berlin, Germany

Email: komnhnoy@auth.gr

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Research in III-nitrides is recently focused in $\text{In}_x\text{Al}_{1-x}\text{N}$ alloys since it was found to be lattice matched to GaN when $x \sim 18$ offering a variety of novel optoelectronic applications. Open issues regarding the electronic properties of InAlN still remain, mainly due to the difficulty of understanding indium diffusion and clustering effects. Scanning transmission electron microscopy combined with Electron energy loss spectroscopy (EELS) is an indispensable tool for studying the structural properties and electronic characteristics of these materials.

$\text{In}_{0.24}\text{Al}_{0.76}\text{N}$ thin films, were grown on GaN/Al$_2$O$_3$ polar templates by metal-organic vapor phase epitaxy (MOVPE) and studied by high resolution transmission electron microscopy (HRTEM) and high resolution scanning electron spectroscopy (HR-STEM) techniques. As it was shown the films were dominated by V-shaped defects terminating at the (0001) sample surface and bound by \{10\overline{1}1\} sidewall facets. V-defects were associated with threading dislocations with large Burgers vectors and their density was equal to $7.5 \times 10^9 \text{ cm}^{-2}$ [1]. In this work we report results derived from the analysis of EELS experimental spectra, recorded from V-defected areas of the $\text{In}_{0.24}\text{Al}_{0.76}\text{N}$ thin films, which were modelled using computational techniques.

In the HR-STEM mode, EELS line scans were taken across the sidewall facets and the core of V-defects (Fig.1). Electron energy loss near edge structure (ELNES) spectra were attained including the carbon C-K, nitrogen N-K, indium In-M$_{4,5}$ and oxygen O-K edges. Intensity changes detected in the spectra recorded in the core, the sidewall facets and along the traces of the sidewall facet junctions, revealed indium fluctuations. Moreover, the observed changes of the shape of the N-K edge indicated different chemical bonding environment. Structural models were developed in order to clarify possible oxygen contamination and indium compositional fluctuations in these defects.

The electronic properties and the chemical bonding of AlN, $\text{In}_x\text{Al}_{1-x}\text{N}$ and $\text{AlN}_{1-y}\text{O}_y$ alloys ($x, y=0.0625, 0.125, 0.25$) were studied using Density functional theory (DFT) calculations as implemented in the WIEN2k code and the TELNES.2 program [2]. The quasi-ternary InAlNO alloy was also studied to scrutinize the ELNES spectra and investigate the simultaneous presence of In-N, Al-O and In-O bonds (Fig.2). In all cases examined the lattice constants of the structural models were optimized. The parameters used to study the bonding environment are the broadening of the edges, the intensity changes and the energy shifts. The N-K edge, that represents the excitations of the N 1s valence electrons to the empty N 2p states of the conduction band (1s$\rightarrow$2p), has been found to be highly sensitive to the concentration as well as the bonding environment of indium as well as oxygen atoms. The ELNES spectra of the N-K edge revealed that the concentration of oxygen impurities affects the edges’ intensity but does not affect their positions. Contrary, indium content in InAlN is predicted to change both the position and broadening of the edges. Lower indium concentrations were identified by calculations on the traces of sidewall facet junctions of the V-defect since the contribution of the AlN matrix beneath the In-rich edges, affecting the experimental spectra, was not taken into account. Based on the above, the interpretation of the experimental spectra led to the conclusion that AlN is dominant at the V-defect sidewall facets while indium and oxygen migrate along the defect core and the sidewall edges.

References

Figure 1. HR-STEM image of the V-defect in InAlN, the green lines (left) that indicate the path of the EELS line scans (right) from the center of the V-defect and between the sidewall facets.

Figure 2. ELNES spectra of the N-K edge of InAlN, AlNO and InAlNO structures. One indium atom in the AlN matrix minimizes the second edge (409eV), broadens the edge by moving the third edge at 411eV, but no changes in relative intensities are observed. One oxygen atom minimizes the intensity of the first edge (405eV). When In-O bonding is present, only the 424eV edge is shifted. When Al-O and In-N bonds are present, indium increases the intensity of the 405eV edge and broadens the N-K by moving the last edge to 427eV.