Direct Imaging of In-rich Nanoclusters in InGaN Correlating Highly Spatially Resolved Cathodoluminescence Microscopy, Transmission Electron Microscopy and Micro-Raman Spectroscopy

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InGaN is the most prominent candidate for the active region of blue light emitting diodes and in particular laser diodes. Severe problems arise from the fact, that InGaN is neither lattice matched to any common substrate, nor to the GaN confinement layers involved in such a device. A complex crystalline nanostructure of inhomogene chemical composition arises in the ternary InGaN epilayers. This creates a strong spatial localization of the electrons, holes, and excitons. Even if this localization isn't strong enough for quantum confinement (i.e., no In rich Quantum Dots are formed), a strong impact on the recombination kinetics is expected: the localized carriers are protected from non-radiative recombination centers [illegible] dislocations, ...). A detailed microscopical characterization yielding [illegible] structural, chemical and optical properties on a nanometer scale [illegible] physical understanding of the fundamental recombination processes.
Here we present comprehensive investigation by highly spatially resolved (lateral resolution < 50nm) and time resolved cathodoluminescence microscopy (CL), micro-Raman Spectroscopy (μ-Raman) in correlation with analytical and high-resolution transmission electron microscopy (TEM), energy-dispersive X-ray microanalysis (EDX) and atomic force microscopy (AFM). The sample under study is a 1μm thick InGaN layer with an average In-content of [In]average = 4.5%, grown by MBE on basal plane sapphire using a 30nm thick GaN buffer.
Plan view and cross sectional mappings of the local CL emission wavelength visualize lateral and vertical inhomogeneities in the chemical composition of InG[illegible] an overall vertical In-gradient ([In] decreases in c-direction) and an In accumulation close to the surface, local In rich nano-clusters showing a columnar structure in growth direction are observed. These results quantitatively agree with mappings of In to Ga ratio obtained by EDX in TEM. The overall In concentration decreases slowly in growth direction, but there is a thin In rich surface layer. The lateral variations in In concentration are irregular and more pronounced. Vertical columnar domains of high local In-content up to [In] = 30% are identified.
While the laterally averaged CL luminescence shows on very broad band (FWHM = 380meV), local spot mode CL consists of narrow peaks (FWHM < 60meV), showing a strong variation of peak position. A statistical analysis yields no simple mono-modal distribution function but proves a clear preference for discrete CL wavelength, i.e. preference for certain discrete In-concentrations.
Time resolved cathodoluminescence visualizes the thermalization of the excess carriers laterally into the nano-domains of lowest bandgap, i.e. highest In-content.
The surface morphology is imaged by showing the cluster structure with typical sizes of 200nm in quantitative agreement with the CL- and EDX-microscopy.
μ-Raman measurements were carried out with a lateral spatial resolution better than 1μm mapping the local modulation in strain and local free carrier concentration.