Probing Local Stoichiometry in InGaN based Quantum Wells of solid-state LEDs

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It is technologically desirable to improve the efficiency (brightness) of green light emitting diodes (LED) to reach values comparable to other LEDs of the visible spectrum for general lighting applications. The active region of green LED devices consists of In\textsubscript{x}Ga\textsubscript{1-x}N quantum wells (QWs) sandwiched between a p- and n-doped GaN matrix. Unlike all other semiconductor LEDs, engineering of the indium (In) distribution towards inhomogeneities seems to boost device performance. The small difference of atomic numbers between Ga and In atoms ($\Delta Z = 18$), the low absolute number of In atoms in 2-3 nm wide quantum wells of concentration $x \sim 0.2$, and the small size of clusters generate unusual challenges for a quantitative investigation of the relation between structure, chemistry, and optical performance since single atom sensitivity and the application of different analytical tools are of importance. We address this challenge by application of high-resolution TEM (HRTEM), annular dark field (ADF) TEM, and cathodoluminescence (CL).

Samples are prepared site-specific from characterized devices by a focused ion beam (FIB) process followed by wet etching [1]. Quantitative measurements of the local indium concentration at the atomic scale utilize HRTEM. From cross-section images, displacement fields and strain (shown in Fig. 1) can be determined with a resolution of 0.5 x 0.3 nm\textsuperscript{2}. A resolution around 0.1 nm can be obtained by a quantitative intensity interpretation. The distribution of the In rich sites is studied in cross-section and in plan-view (see Fig. 2.).

ADF-TEM [2] provides a positive chemical identification of In clusters in the QW’s. The technique utilizes an annular dark field aperture in the back focal plane of the objective lens allowing to record only electrons scattered through a semi angle of 20-40 mrad [3]. Experimentally, we find that single atom scattering cross sections for GaN measured with this technique are close to Rutherford scattering values (Fig. 3). The chemical sensitivity of the technique is illustrated in Fig. 4, which shows a bright field and a corresponding ADF image of a single InGaN QW. Despite an expected delocalization (due to a finite spherical aberration) In rich clusters can be distinguished.

Room temperature high-resolution CL spectrums are recorded in a SEM both from cross-section and plan-view samples. An example of a plan-view spectrum obtained from a single InGaN quantum well at a magnification of 600kx is shown in Fig. 5. This combination of techniques allows correlating the local (defect) structure and stoichiometry to variation of optical properties and device efficiencies that will be discussed.

References
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FIG 1. HRTEM cross-section image of a single InGaN QW inside a GaN matrix showing local lattice distortions (marked by arrows). Strain measurements (shown as an inset) were taken from the area marked by the white rectangle.

FIG 2. HRTEM plan-view image of a single InGaN QW covered by a few nm thin GaN cap layer. Arrows mark areas with higher contrast of the atomic columns which indicates the presence of the defect like structures as found in cross-section.

FIG 3. Annular dark field (ADF) signal vs. GaN sample thickness. The ADF signal can be described through an electron scattering process that depends exponentially on sample thickness, allowing to determine the electron scattering cross section.

FIG 4a) Bright field image of a single InGaN QW; 4b) Corresponding ADF TEM image. The contrast in the image is chemically sensitive and shows higher brightness for higher In concentration.

FIG 5. Room temperature high-resolution cathodoluminescence (CL) spectrum from a single InGaN quantum well (taken from a plan view sample with thin GaN cap layer)