Growth of Fully Relaxed (In,Ga)N Pseudo-Substrates by a Two-Step Protocol Without Ex-situ Patterning

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The band gap of (In,Ga)N spans the entire visible spectrum. This semiconductor is thus a promising candidate for the fabrication of red-green-blue light emitting diodes (LEDs) from one and the same material class. Blue and green (In,Ga)N LEDs have been commercialized, but the quantum efficiency of red (In,Ga)N LEDs is insufficient for practical purposes. This limitation arises from the large lattice mismatch between the active region with increased In content of around 40% and GaN. The large strain in the QW results in large internal fields, reducing the recombination rate. Also, certain point defects (such as Ca) become increasingly active in nonradiative recombination. We note that in general bulk group-III nitride substrates are expensive, and the long-standing success of blue (In,Ga)N LEDs is a result of mature procedures for the growth of GaN template layers on sapphire and SiC substrates.

In this study, we present a simple yet effective two-step growth protocol without patterning that results in essentially fully relaxed (In,Ga)N pseudo-substrates on GaN templates. The key concept is to grow by plasma-assisted molecular beam epitaxy first a rough (In,Ga)N layer under N-rich conditions and second proceed with overgrowth under metal-rich conditions that leads to a smooth surface.

In detail, the first (In,Ga)N layer is grown at 500 °C under N-rich conditions for 30 min. These growth conditions lead to a spotty reflection high energy electron diffraction (RHEED) pattern typical for a rough surface. The In content and degree of strain relaxation are determined from x-ray diffraction reciprocal space maps (RSM). The In content varies between samples in the range 25 to 42%, and the relaxation degree reaches up to 95%. The thickness of the layer with In content 42% and relaxation degree 95% as measured by scanning electron microscopy is about 200 nm. Characterization by atomic force microscopy (AFM) reveals a surface with a root mean square (RMS) roughness of 4.4 nm in an area of $2\times2\ \mu m^2$.

That N-rich growth leads to a rough surface for group-III nitrides is well known. The main innovation of this study is the demonstration that the surface can be smoothened again in the case of (In,Ga)N. To this end, (In,Ga)N overgrowth is carried out at 550 °C under metal-rich conditions for 60 min. The RHEED pattern exhibits streaks with undulations in intensity, and AFM analysis shows a surface morphology with a clearly reduced RMS roughness of 1.8 nm. These properties are achieved for an In content of ≈30% and a strain relaxation degree of ≈80%. A layer with a lower In content of 24% and similar relaxation degree and surface roughness is obtained by reducing the In flux during growth and increasing the substrate temperature. The photoluminescence spectra acquired at room temperature exhibit a single emission band centered at 574 nm and 543 nm, respectively. The full widths at half maximum (FWHM) are 53 nm (225 meV) and 28 nm (118 meV), respectively. The latter value indicates excellent homogeneity, and the increase compared to a reference (In,Ga)N sample emitting in the blue spectral range and grown directly on a GaN template is related to more pronounced alloy disorder for higher In content.

In conclusion, we have demonstrated the growth of (In,Ga)N layers with high In content up to 42%, smooth surface, and essentially full strain relaxation. These properties stand out in comparison to other approaches for the fabrication of pseudo-substrates, and the facile two-step growth protocol can easily be scaled up. Therefore, this advancement represents a critical step toward the realization of high-performance red (In,Ga)N LEDs for next-generation micro-LED applications.

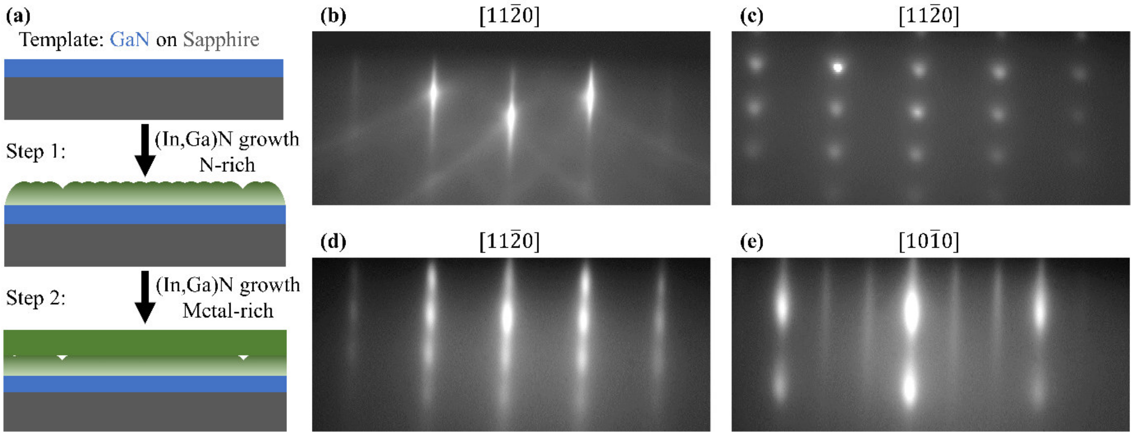

Figure 1: (a) Schematic of the two-step growth protocol for the fabrication of relaxed (In,Ga)N pseudo-substrates. RHEED patterns of (b) the GaN(0001) template along the $[11\bar{2}0]$ direction, (c) the first (In,Ga)N layer along the $[11\bar{2}0]$ direction, and the second (In,Ga)N layer along the (d) $[11\bar{2}0]$ and (e) $[10\bar{1}0]$ directions. The spotty and streaky patterns show the surface smoothening during overgrowth. The $(\sqrt{3} \times \sqrt{3})$R30° surface reconstruction seen in (e) reflects that growth was carried out with an In adlayer at the growth front.

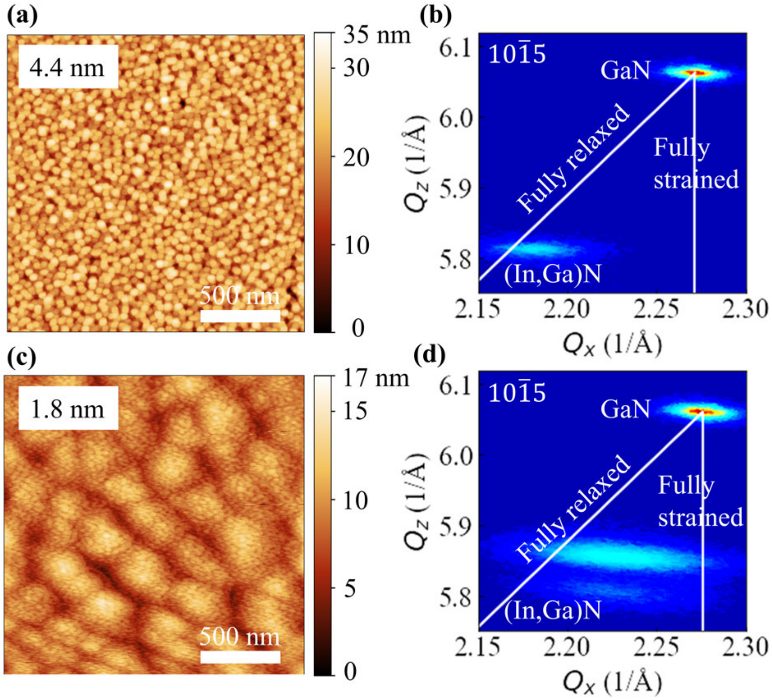

Figure 2: (a) AFM topograph of the first (In,Ga)N layer with a RMS roughness of 4.4 nm. (b) RSM of the first (In,Ga)N layer around the 105 reflection revealing a strain relaxation degree of 95% and In content of 42%. (c) AFM topograph of the second (In,Ga)N layer with a RMS roughness of 1.8 nm. (b) RSM of both the first and second (In,Ga)N layer around the $10\bar{1}5$ reflection indicating a strain relaxation degree of 80% and In content of 30%.

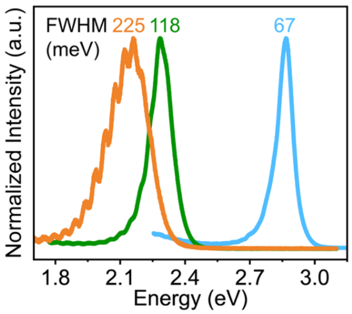

Figure 3: Photoluminescence spectra of three (In,Ga)N samples acquired at room temperature. The blue curve corresponds to a 500-nm-thick reference (In,Ga)N layer with In content of 13% grown directly on a GaN template. The green and orange curves correspond to the (In,Ga)N pseudo-substrates grown by the two-step protocol with In contents of 24 and 30% in the second layer, respectively. The text labels indicate the FWHMs of the spectra.