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1 Introduction The narrow band gap of InN has the potential for use in new applications including photonic devices suitable for optical communications. Alloys of InN with Ga and Al may also be used in applications for high efficiency solar cells [1]. Despite this potential, the difficulty of obtaining high quality material constitutes one of the reasons for the slow device development. In this paper we report on the growth of high quality InN on a production style, molecular beam epitaxy system using an RF plasma.

2 Experimental The growth was performed using two-step method on 2 inch MOCVD GaN templates. A GEN200® plasma assisted molecular beam epitaxy (PA-MBE) system, equipped with multi-wafer (13x2 in) platens and adapted for autonomous operation, was used for the growth of GaN and InN layers. High purity N_2 flow has been used with a Veeco UNI-Bulb® RF plasma source to deliver the active nitrogen to the growth surface. The substrates have been overgrown at 720 °C with a 300 nm GaN buffer deposited by PA-MBE under conditions known to produce step-flow growth and droplet-free surfaces. Layers of 1μm thick InN were deposited in the temperature range from 450 °C to 500 °C and a growth rate of 0.25 μm/h using a nitrogen flow of 8 sccm and an RF power of 600 W under slightly In-rich conditions [2]. Two groups, A and B, of InN samples were grown using different growth methodologies, detailed bellow.

3 Results and discussion For group A, the growth conditions that have been established at the beginning of the growth were maintained unchanged for the entire deposition period. For this group, the 2-dimensional (2D) RHEED pattern at the beginning of the growth underwent a transition to 3-dimensional (3D) growth mode to various degrees for all but one sample. As a result of this transition, the samples of group A exhibit rough surfaces. This 2D to 3D growth evolution is indicative of small deviations from the optimal III-V ratio on the growth surface. The adaptive deposition method used for group B, provides the ability to adjust the III-N ratio on the growth surface within a narrow range. For this method the group III effusion cell shutter is actuated ON and OFF based on the RHEED pattern intensity. As a result a streaky RHEED pattern indicative of a 2-dimensional surface is maintained throughout the growth period.

The AFM analysis of the InN layers of group B revealed a smooth surface with monolayer height teraces (0.281 nm), characteristic of the step-flow growth mode. The measurement indicates a root mean square (RMS) roughness of 0.7 nm for the (2x2) µm^2 scan and 1.4 nm for the (5x5) µm^2 scan. In Fig. 1 the surface morphology of a (2x2) µm^2 area is presented.

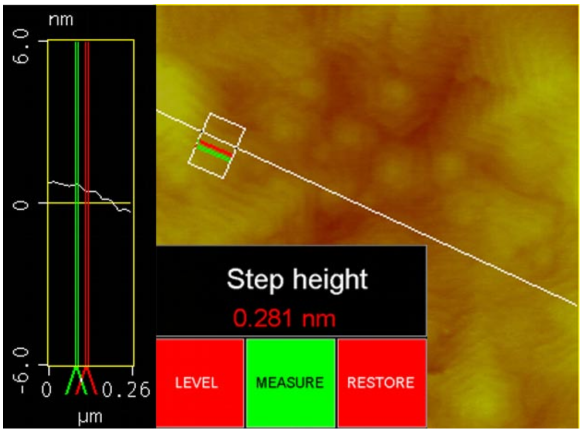

Figure 1 InN film showing step-flow characteristics.

This surface characteristics are similar to the recently-reported step-flow growth of In-polar InN by MBE [3].

Under ideal conditions the formation of InN epitaxial surface, takes place under a uniform adlayer of In with a thickness of no more than few monolayers. The vapor-liquid-solid (VLS) growth mode is the mechanism that allows active nitrogen radicals to permeate In adlayer, and reach the epitaxial layer.

Under less than ideal conditions, In droplets may form during the early stages of the InN growth and they are likely to be sustained throughout the growth. Chemical removal of the In droplets uncovers a surface populated by small InN crystals with shapes that are reminiscent of hexagonal symmetry. We conjecture that within the In droplet, the average length of the bulk diffusion of the active nitrogen may be shorter than the radius of the droplet. Thus nitrogen would either recombine to form neutral molecular nitrogen or, more likely, would form InN slabs in suspension. As a result, the epitaxial surface would be depleted of nitrogen species and the growth will be retarded, resulting in substantially different crystal formation and surface morphology. The SEM picture in Fig. 2 shows such InN crystals in suspension and in the insert the morphology of the defective area underneath the droplet can be seen.

Crystal quality has been investigated by XRD, using both symmetric and asymmetric diffraction arrangements.

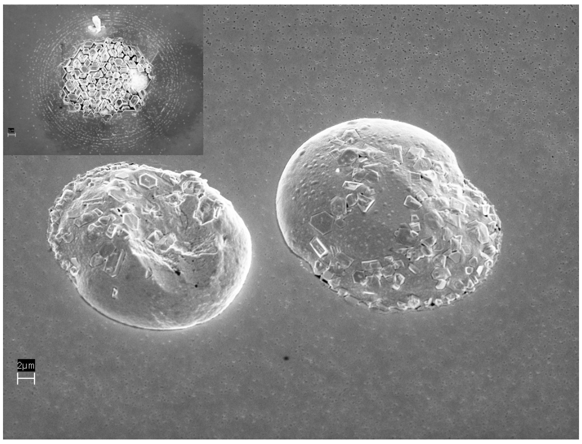

Figure 2 In droplet with crystals in suspension.
Insert: Defective area after In droplet removal.

Threading dislocations (screw and edge) are the dominant defects present in InN epitaxial layers [4]. Screw dislocation densities in the range of (2-7) x 10^9 cm^{-2} were calculated from the FWHM of (0002) reflection. Also the XRD estimated edge dislocation densities are in the range of (6-13) x 10^{10} cm^{-2}.

The effectiveness of growth temperature as a factor in the reduction of the number of threading dislocations has been investigated. For our growth conditions we have found that a minimum of dislocation density can be obtained for a growth temperature of ~ 475 °C as depicted in Fig. 3.

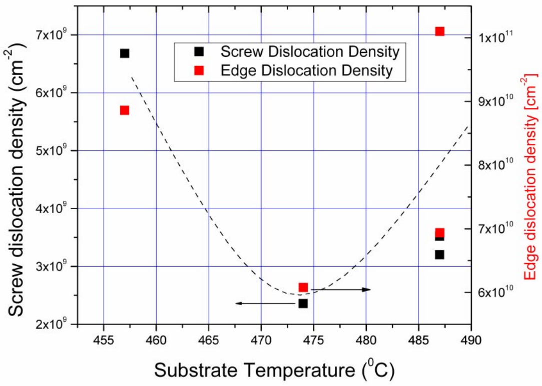

Figure 3 Growth temperature effect on the dislocation density. Dashed line is guide for eye only.

The (0002) ω-scan and 2θ/ω-scan measurements have been performed on both MOCVD GaN templates and InN layers. For the GaN templates, (0002) ω-scan measurements showed FWHM in the range 1150±20 arcsec, while InN grown on these templates exhibits slightly larger FWHM values. Representative 2θ-ω scan for a typical InN epilayer are presented in Fig. 4.

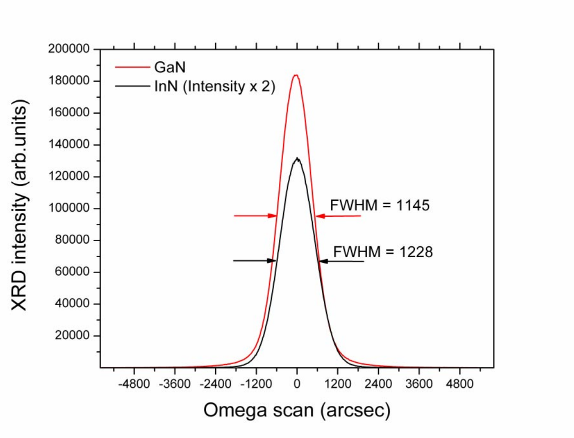

Figure 4 ω-scan of GaN template and InN layer.

Clearly seen are narrow diffraction lines from the GaN template and the overgrown InN layer.

Hall measurements at room temperature in the van der Pauw geometry have been performed for all samples. For the samples in group A the mobility was in the range from 1024 cm^2/Vs to 1481 cm^2/Vs. All samples in group B grown under adaptive mode, exhibited higher mobility, with a range from 1404 cm^2/Vs to 1904 cm^2/Vs. The sample with the highest room-temperature mobility, exhibited a mobility of 2106 cm^2/Vs at 80K. This higher mobility at low temperature is consistent with previously published measurements of high-quality InN [5]. Samples which exhibited step-flow growth features, had comparable mobilities with values slightly higher than 1400 cm^2/Vs, and average electron concentration of ~ 3.2 ×10^{18} cm^{-3}.

The analysis of sample-to-sample differences in free carrier concentration do not suggest a relation to screw or edge dislocation densities.

Raman spectra have been measured for all InN samples in this study. The symmetry allowed E_2^2 and A_1(LO) phonons with energies corresponding to 493.0 and 584.6 cm^{-1} dominate the spectra at room temperature.

Upon cooling to 20 K, the phonons blue shift by 1.9 cm^{-1} for E_2^2 and 4.9 cm^{-1} for A_1(LO). The E_2^2 phonon exhibits very narrow line width, ~ 4.2 cm^{-1} at ambient temperature, comparing well with previously published results, and indicative of high crystal quality [6].

For the same InN samples, RT photoluminescence has been measured, using an InGaAs detector and an Ar$^+$ laser at 514 nm. The PL intensity is strong in each sample described here. A typical PL spectrum for the InN layers is presented in Fig. 5. The samples in group A exhibit a single emission peak at energies between 0.635 eV and 0.658 eV. The band-edge PL is observed to shift systematically toward higher photon energy with increasing free-carrier density. The measured shift is ~23 meV as the carrier concentration increases from 6×10^{17} to 4×10^{18} cm^{-3}. The shift observed in the band edge with free carrier density has been previously attributed to Moss-Burstein effect [7].

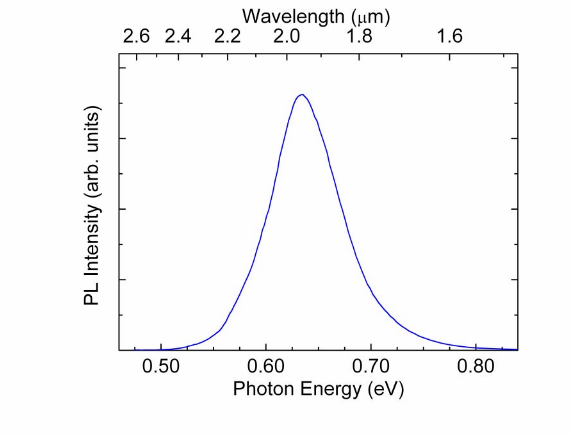

Figure 5 Typical photoluminescence of the InN layers.

4 Conclusion In conclusion, we have demonstrated the step-flow growth mode, with monolayer height terrace steps, using a production-style PA-MBE system, GEN200®. We have also observed the consequences of unintentional In droplets formation during the growth. We propose VLS as the growth mechanism under which the droplet formation impedes InN growth. The room temperature Hall mobility of InN layer, grown in such step-flow mode is slightly higher than 1400 cm^2/Vs while for growth conditions yielding a smooth surface with no well-defined steps, mobility as high as 1904 cm^2/Vs at room temperature has been obtained. Raman spectra show very narrow E_2^2 lines indicative of excellent InN crystalline quality. The samples have intense PL emission at room temperature in the 0.64 to 0.66 eV range. The weak shift is consistent with free-carrier concentration increase effects.

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References

[1] J. Wu, W. Walukiewicz, K.M. Yu, W. Shan, J.W. Ager III, E.E. Haller, H. Lu, W.J. Schaff, W.K. Metzger, and S. Kurtz, J. Appl. Phys. **94**(10), (2003).

[2] K. Xu and A. Yoshikawa, Appl. Phys. Lett. **83**(2), 251 (2003).

[3] X. Wang, S.-B. Che, Y. Ishitani, and A. Yoshikawa, Jpn. J. Appl. Phys. **45**(28) (2006).

[4] H. Heinke, V. Kirchner, S. Einfeldt, and D. Hommel, Appl. Phys. Lett. **77**, 2145 (2000).

[5] J.S. Thakur, R. Naik, V.M. Naik, D. Haddad, G.W. Auner, H. Lu, and W.J. Schaff, Mater. Res. Soc. Symp. Proc. **892** (Fall Meeting) (2006).

[6] M. Giehler, M. Ramsteiner, P. Waltereit, O. Brandt, K.H. Ploog, and H. Obloh, J. Appl. Phys. **89,** 3634 (2001).

[7] J. Wu, W. Walukiewicz, W. Shan, K.M. Yu, J.W. Ager III, E.E. Haller, Hai Lu, and W.J. Schaff, Phys. Rev. B **66**, 201403(R) (2002).