In situ Studies of Surface Morphological Evolution During Indium Nitride Growth by Atomic Layer Epitaxy

C.R. Eddy, Jr.[1], N. Nepal[1], S.G. Rosenberg[2], V.R. Anderson[3], J.M. Woodward[2], C. Wagenbach[4], A.C. Kozen[2], Z.R. Robinson[5], L.O. Nyakiti[6], S.B. Qadri[1], M.J. Mehl[7], K.F. Ludwig[4] and J. K. Hite[1]

[1]U.S. Naval Research Laboratory, Washington, DC,
[2]ASEE, Washington, DC (residing at NRL),
[3]Sotera Defense Solutions, Lanham, MD,
[4]Boston University, Boston, MA,
[5]SUNY Brockport, Brockport, NY,
[6]Texas A&M University, College Station, TX ,
[7]U.S. Naval Academy, Annapolis, MD

Nitride semiconductors have had significant commercial success, but full bandgap engineering of these materials is prohibited by the high temperatures used in conventional growth methods. Recently, we have developed a growth method –low temperature atomic layer epitaxy (ALEp) – that has empirically produced crystalline semiconductor films with properties comparable to those from conventional growth processes, but at roughly half the growth temperature [1,2]. This has eliminated miscibility gaps in ternary III-N semiconductor films and enabled the realization of full bandgap engineering from 0.7 eV to 6.1 eV.

Despite these empirical successes, the fundamental mechanisms involved in ALEp are unknown and the full promise of the method unrealized. To obtain such enabling knowledge we have employed synchrotron-based grazing incidence small angle x-ray scattering (GISAXS) to study the low temperature atomic level processing (ALP) of GaN substrate surfaces for epitaxy and ALEp nucleation and growth of InN on said surfaces. GISAXS allows real-time, in situ monitoring of the surface morphology during these processes.

In this presentation, we will introduce the GISAXS method and the apparatus we have developed to conduct in situ GISAXS measurements of the aforementioned ALP and ALEp processes. We monitor the evolution of GaN substrate surface morphology during a series of low-temperature ALPs including: gallium flash off (GFO), hydrogen plasma clean, and nitrogen plasma nitridation. We learn that the optimum surface results from a GFO conducted at 500°C for only 10 cycles followed by a hydrogen plasma clean. Further, we learn that conventional plasma nitridations are detrimental to smooth surface evolution. When employed to study ALEp InN nucleation and growth, GISAXS data, coupled with Porod[3] and 2D Fourier Transform analysis, affords a clear picture that the growth proceeds by island nucleation and growth and not by the conventionally accepted layer-by-layer growth associated with atomic layer deposition. We have monitored the evolution of island nucleation density, island spacing, island shape and island size as a function of key ALEp growth parameters. We observe that the islands are generally tens of nm or less in size and evolve from a spherical mound shape to a cylindrical shape. Finally, we will present the variations between 2D and 3D growth modes with growth parameter variations that provide insights on process modifications to promote higher quality electronic materials growth.

1. Nepal et al., Cryst. Growth and Des., 13, 1485 (2013)
2. Nepal et al., Thin Solid Films 589, 47 (2015).
3. G. Porod, Kolloid Z., 124, 83 (1951).

In situ Studies of Surface Morphological Evolution During Indium Nitride Growth by Atomic Layer Epitaxy

C. R. Eddy, Jr. et al., U. S. Naval Research Laboratory

Supplemental Information

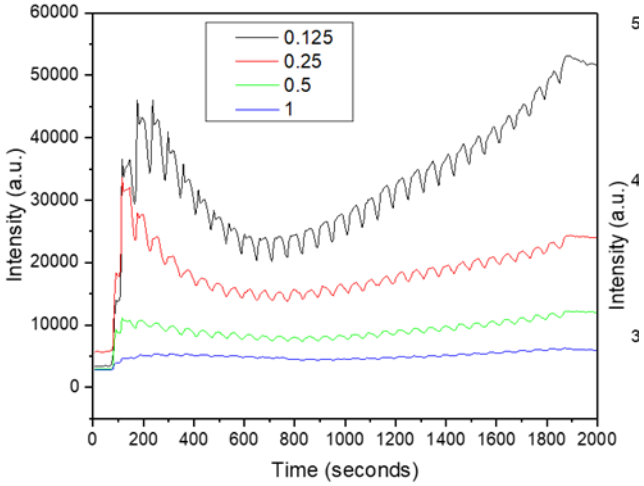

Fig. 1. Evolution of GISAXS scattering intensity at various length scales during an emulated gallium flash off atomic level processing of GaN substrate surfaces at 500°C. ALP cycles are evident as is a minimum in scattering after 10 cycles.

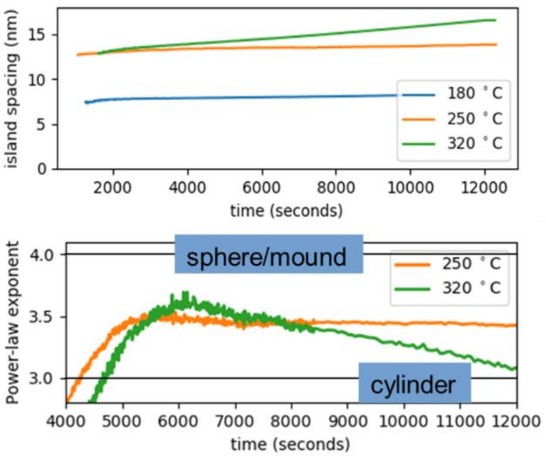

Fig. 2. Evolution of island spacing and island shape during InN growth on optimally prepared GaN substrate surfaces at temperatures between 180°C and 320°C. Clear coarsening of island size is observed at 320°C as is a transition of island shape from hemispherical mounds to cylinders.

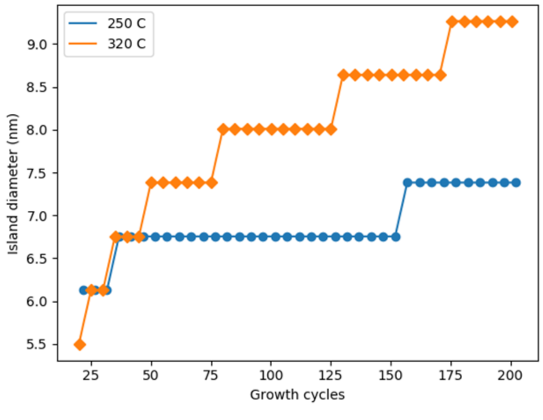

Fig. 3. Evolution of InN island diameter with growth time at 250 and 320°C.   Island diameter derived from Hankel transformation of GISAXS scattering data. Behavior suggests more two-dimensional growth at 320°C.

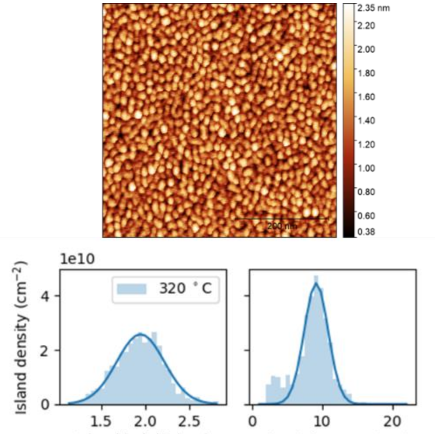

Fig. 4. Post growth atomic force micrograph and analysis of InN film grown on GaN substrates at 320°C confirming island diameter and elucidating the island height.