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# Radiation Effects and Defects in Solids: Incorporating Plasma Science and Plasma Technology

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# Ion beam-mixing effects in nearly lattice-matched AllnN/GaN heterostructures by swift heavy ion irradiation

G. Devaraju  $^{\rm a}$  , S. V.S. Nageswara Rao  $^{\rm a}$  , N. Srinivasa Rao  $^{\rm a}$  , V. Saikiran  $^{\rm a}$  , T. K. Chan  $^{\rm b}$  , T. Osipowicz  $^{\rm b}$  , M. B.H. Breese  $^{\rm b}$  & A. P. Pathak  $^{\rm a}$ 

<sup>a</sup> School of Physics, University of Hyderabad, Central University P.O, Hyderabad, 500 046, India

<sup>b</sup> Department of Physics, Centre for Ion Beam Applications (CIBA), National University of Singapore, Singapore, 117542, Singapore

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# Ion beam-mixing effects in nearly lattice-matched AlInN/GaN heterostructures by swift heavy ion irradiation

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<sup>a</sup>School of Physics, University of Hyderabad, Central University P.O, Hyderabad 500 046, India; <sup>b</sup>Department of Physics, Centre for Ion Beam Applications (CIBA), National University of Singapore, Singapore 117542, Singapore

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Ion beam-induced intermixing is of great interest and has been observed in various systems such as metal–semiconductor interfaces. However, similar effects in III-nitrides have not been studied in any detail, yet. In the present study, swift heavy ion-induced intermixing of nearly lattice-matched  $Al_{(1-x)}In_xN/GaN$  heterostructures have been investigated using Rutherford backscattering spectrometry and high-resolution X-ray diffraction techniques. Inter-diffusion of Ga and In elements across the  $Al_{(1-x)}In_xN/GaN$  interface and the consequent formation of a new mixed quaternary alloy (AlGaInN) layer have been observed. The influence of electronic energy loss of SHI on intermixing effects has been studied.

Keywords: III-nitrides; swift heavy ion irradiation; RBS;HRXRD

## Introduction

The family of III-nitrides has numerous applications in optoelectronic, high-power and high-frequency devices. In particular, AlGaN and InGaN heterostructures (HS) have been investigated extensively compared to AlInN. On the other hand, high-quality nearly lattice-matched AlInN/GaN HS were reported by several research groups (1-7). These structures have been used for realizing Bragg reflectors, microcavities and transistors (3, 6, 8, 9). Furthermore, AlInN alloys cover extremely wide spectral range from deep UV to infrared. However, the growth of AlInN over the full compositional range is difficult due to large disparities in cation size  $(0.53 \text{ Å for } \text{Al}^{3+} \text{ and } 0.76 \text{ Å for } \text{In}^{3+})$  (9) and thermal properties of the binary constituents (10-12). Recently, it was demonstrated that biaxial strain (which depends on composition and thickness) is vital and influences miscibility gap (12). Hence, it is important to study the various factors that can influence the compositional gradients of these HS.

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<sup>\*</sup>Corresponding author. Email: appsp@uohyd.ernet.in

Owing to their technological importance in satellite communications, there is a need to understand how the device performance degrades/improves under various kinds of irradiations including heavy ion bombardment. Thus, there is a great demand to understand the interface mixing and delimitation of HS under the influence of heavy ion irradiation. It is well known that when a fast moving ion passes through solid materials, it transfers large amounts of energy to the electronic subsystem through inelastic collisions by either electronic excitation or ionization of the target atoms (13). At these velocities, ions in materials can create columnar defects (14), cylindrical tracks (15), amorphization (16) and recrystallizations (17). Most of the high-energy irradiation results were understood by exploiting the concepts of planar defects, delamination and tracks propagation (18, 19).

In the recent past, it has been observed (20) that the electronic energy deposition beyond critical value can cause the movements of atoms leading to intermixing of multilayers. Swift heavy ion-(SHI) induced mixing of various metal–semiconductor and semiconductor–semiconductor interfaces has been studied by several groups (20–26). The amount of mixing is found to be dependent on the amount of energy that is transferred to target electrons. It is observed that the intermixing is a result of diffusivity of atomic species during the transient molten phase in picoseconds range. However, similar effects in III-nitrides have not been studied in any detail yet. Here, we study SHI-induced mixing of nearly lattice-matched  $Al_{(1-x)}In_xN/GaN$  HS. Compositional profiles of as-deposited and irradiated samples have been studied using Rutherford backscattering spectrometry (RBS) and high-resolution X-ray diffraction (HRXRD) techniques. Inter-diffusion of Ga and In elements across the  $Al_{(1-x)}In_xN/GaN$  interface and the consequent formation of a new mixed quaternary alloy ( $Al_xGa_yIn_{1-x-y}N$ ) layer have been observed. The influence of electronic energy loss of SHI on intermixing effects has also been studied.

#### **Experimental details**

 $Al_{(1-x)}In_xN/GaN$  HS were grown on (0001) a sapphire substrate by the metal organic chemical vapour deposition technique. These samples were then irradiated by 70 MeV Ni and 100 MeV Ag ions at a fixed fluence of  $1 \times 10^{12}$ /cm<sup>2</sup> using the 15 MV Pelletron Accelerator (22) at Inter-University Accelerator Centre (IUAC), New Delhi, India. Pristine, 70 MeV Ni and 100 MeV Ag ions irradiated samples have been named as T2A, T2B and T2C, respectively. The purpose of this experiment is to study the influence of electronic energy loss of SHI on  $Al_{(1-x)}In_xN/GaN$ HS at a fixed fluence. Electronic energy loss ( $S_e$ ) and nuclear energy loss ( $S_n$ ) of 70 MeV Ni ions and 100 MeV Ag ions in HS have been calculated using stopping and range of ions in matter code (SRIM 2003). Thus,  $S_e$  and  $S_n$  for 70 MeV Ni ions in these HS have been found to be 10 and 0.02 keV/nm, similarly for 100 MeV Ag ions in HS it has been found to be 16 and 0.08 keV/nm, respectively. Corresponding projected ranges of Ni and Ag ions in these HS are 11.24 and 11.13 µm, respectively. Thus, ions predominantly deposit energy to target electrons uniformly throughout the thin film which is two orders of magnitude smaller compared to the projected range. RBS studies have been performed with a collimated beam of 2 MeV He<sup>+</sup> ions using a 2.5 MV model AN-2500 Van-de-Graaff accelerator (High Voltage Engineering) at National University of Singapore (NUS), Singapore. The backscattered He<sup>+</sup> ions have been detected at an angle of 170° using a silicon surface barrier detector with an energy resolution of 15 keV. We have used the SIMNRA simulation code for fitting the measured RBS spectra. The structural properties of pristine and irradiated AlInN/GaN HS have also been characterized by HRXRD. These measurements around (0002) GaN reflection are carried out by a Bruker D8 DISCOVER, X-ray diffractometer with  $CuK_{\alpha}$  as the source and a four-bounce Ge (220) monochromator.

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### **Results and discussion**

RBS measurements were performed on pristine and irradiated samples (T2A, T2B and T2C) for determining Ga and In compositional profiles. The presence of In, Ga and Al can be observed in the RBS spectrum of the pristine sample (T2A) as shown in Figure 1. Simulations were carried out using the SIMNRA code by varying the layer parameters starting with nominal values until a satisfactory fit is obtained. These measurements suggest that the presence of Ga in the AlInN layer may be due to a possible diffusion of Ga and In across the interface during the growth process. The average In composition is estimated to be around 21% in this layer as against the nominal value of 18%. Similar compositional gradients have been reported earlier for AlInN films (23, 24). Generally, AlInN films close to lattice-matched conditions show pseudomorphic growth (24). In the present study, the observed compositional gradient may lead to a slight lattice mismatch between the AlInN layer and the GaN substrate. This is further confirmed by the HRXRD measurements. Symmetric rocking curve measurements were performed along the (0002) axis of GaN for determining the average In composition and lattice strain. Figure 2 shows the measured and simulated  $\omega - 2\theta$  scans of the pristine sample (T2A). HRXRD simulation is carried out using the dynamical theory-based Philips Epitaxy software. The composition and thickness of the layer is optimized by a trial-and-error method, starting from nominal values until a satisfactory fit is observed. The simulated scan matches reasonably well with experimental data. The average In composition and the lattice parameter (c) obtained from these fits are given in Table 1. It may be noticed that RBS in irradiated samples yields a gradient of In composition with depth. These values can be used for evaluating the out-of-plane strain in the AlInN layer.

The measured RBS and HRXRD spectra of pristine and irradiated samples (T2A, T2B and T2C) are shown in Figures 3 and 4. These spectra confirm the SHI-induced diffusion of In and Ga elements across the AlInN/GaN interface. The enhanced diffusion of In and Ga due to 100 MeV Ag ion irradiation in T2C sample demonstrates the influence of  $S_e$  on SHI-mixing effects. The Ga-edge from GaN substrate in RBS spectra is found to be at lower energies in irradiated samples, particularly in T2C, as compared to pristine sample. This further confirms the formation of a thin



Figure 1. Measured and simulated RBS spectra of the T2A sample showing Ga diffusion into the indium energy window.



Figure 2. (0002)  $\omega - 2\theta$  X-ray diffraction for the T2A sample along with the theoretical simulated profile.

Table 1. Average indium composition of T2A, T2B and T2C samples obtained from HRXRD and RBS measurements.

Sample ID	Measured lattice parameters (by HRXRD) C (Å)	Average indium composition by	
		HRXRD	RBS
T2A (Pristine)	5.150	0.21	0.21
T2B (Ni)	5.140, 5.212 (bi layer)	0.20, 0.29	Gradient
T2C (Ag)	5.146, 5.205 (bi layer)	0.21, 0.28	Gradient



Figure 3. RBS spectra of T2A, T2B and T2C samples. SHI-induced diffusion of Ga from GaN into the AlInN epilayer is evident in spectra.



Figure 4. The (0002) HRXRD rocking curves of T2A, T2B and T2C samples. SHI-induced modification of strain in the epilayer is evident in spectra.

intermixed layer between the AlInN layer and the GaN substrate. In fact, SIMNRA simulation of the T2C sample suggested the formation of an interdiffused layer (InGaAlN) with concentration gradients between InAlN and GaN layers.

The influence of SHI-induced mixing on the lattice strain in AlInN layer is studied using HRXRD measurements. Measured values of In composition and corresponding lattice constants are given in Table 1. Figure 4 clearly shows a significant shift in the Bragg angle of layer peak and a corresponding change in strain due to the SHI irradiation as a function of  $S_e$ . It is important to note that the ion irradiation can be used to alter the strain by varying  $S_e$ . In fact, tensile strain is induced (T2C) in an initially nearly lattice-matched/compressive strained layer (T2A).

It is known that ion beams at moderate fluence can create latent tracks in various metal– semiconductor and semiconductor–semiconductor bi layer systems (20). Such SHI-induced intermixing has been observed by different research groups in different systems such as Fe/Si, C/Si, C<sub>60</sub>/Si, Co/Si, V/Si, Zr/Si, Cu/Ge, Ni/Si and Fe/Ni multilayers. In all the above studies, SHI-induced mixing at the interfaces has been identified as atomic diffusion in the molten phase created by transient temperature spike (25, 26). In the present study, It is noted from Figures 3 and 4 that T2C sample shows more intermixing than T2B sample, which is due to a significant difference in S<sub>e</sub> deposition rates for their corresponding ion propagation in Al<sub>(1-x)</sub>In<sub>x</sub>N/GaN HS. The enhanced diffusion of In and Ga across the AlInN/GaN interface at higher S<sub>e</sub> (*i.e.* in T2C) can be understood as a consequence of high quenching rates along the path of ions.

### Conclusions

SHI-induced mixing effects in nearly lattice-matched  $Al_{(1-x)}In_xN/GaN$  HS have been studied as a function of  $S_e$ . The RBS technique has been used for determining composition profiles as a function of depth on pristine and irradiated samples. Compositional gradients have been observed in the measured RBS spectrum of the pristine sample. Average compositions obtained from RBS and HRXRD are in close agreement with nominal values. Moreover, the observed diffusion gradient in RBS and HRXRD measurements is correlated with simulations. Change of strain status from nearly lattice-matched/compressive strain to tensile strain has also been observed as function of  $S_e$ . Here, it is shown that the SHI-mixing can be used to alter the strain in AlInN/GaN HS. The

present study indicates that it is possible to achieve a new mixed quaternary alloy (AlGaInN) due to interface mixing of AlInN and GaN layers with ion beams. However, it may be quite difficult to achieve by the usual growth methods.

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