Materials Science in Semiconductor Processing 64 (2017) 95–100

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

Tailoring the structural and optical characteristics of InGaN/GaN multilayer thin films by 12 MeV Si ions irradiations

Ishaq Ahmad[a], M. Madhuku[b], Adeela Sadaf[a,c], Shakil Khan[d,*], Javaid Hussain[a], Awais Ali[e], D. Wan[f], S.Z. Ilyas[c], G. Mola[g], Abdul Waheed[h], Muhammad Asim Rasheed[d]

[a] National Center for Physics, Quaid-i-Azam University, Islamabad 44000, Pakistan
[b] iThemba LABS, National Research Foundation, P. Bag X11 WITS 2050, South Africa
[c] Physics Department, Allama Iqbal Open University, Islamabad, Pakistan
[d] Department of Metallurgy and Materials Engineering, Pakistan Institute of Engineering & Applied Sciences (PIEAS), Islamabad 45650, Pakistan
[e] Center for Micro and Nano Devices, Department of Physics, COMSATS Institute of Information Technology, Islamabad, Pakistan
[f] School of Materials Science and Engineering, Shanghai University, Shanghai, China
[g] School Chemistry & Physics, University of Kwazulu-Natal, Pietermaritzburg Campus, Private Bag X01, Scottsville 3209, South Africa
[h] Department of Physics, Islamia College Peshawar (Public sector University), Pakistan

ARTICLE INFO

Keywords:
A. Semiconductor
A. Thin films
B. Ions irradiations
C. X-ray diffraction
D. Raman spectroscopy

ABSTRACT

In this study, the influence of Si ions irradiations (12 MeV energetic) on structural and optical characteristics of InGaN/GaN thin film has been investigated. Irradiation was performed at different Si ions fluences in the range of 1×10^{13} to 1×10^{15} ions/cm^2. X-ray diffraction (XRD) pattern of pristine film indicates only the (0 0 2) oriented crystallites of InGaN while the irradiated films patterns showed other phases (InN and GaN) as well. Ion irradiations at different dose rates have shown no or negligible effect on grain size of InGaN except a shift in the peak position which demonstrates the development of tensile stresses. The existence of other phases in the irradiated films patterns is the indication of InGaN phase separation. Defects produced due to irradiation were also confirmed from peak shifting and appearance of new peak at 669 cm^{-1} in Raman spectra. A decrease in optical bandgap with the increase of ion irradiation dose rate is being reported in this work.

1. Introduction

The ternary $In_{1-x}Ga_xN$ alloy system has attracted much attention in the scientific community due to its unique electrical and optical properties [1]. Its optical bandgap can be tailored in a wide range covering ultraviolet to near-infrared covering over more than 80% of the solar spectrum, depending upon the composition, deposition method, growth parameters and post growth treatments. This makes InGaN an ideal semiconductor for applications in medical photonics and communication devices [2–4]. Therefore, investigations on the effects of composition, strain and phase separation on optical properties offer a wide range of study.

InGaN alloys have numerous useful properties and are different from those of their constituent components, like tailorable bandgap which can lead to the emission of different colors of light. However, under certain conditions, this alloy breaks down into their binary fractions or into regions considerably richer or poorer in one constituent than would be expected from the overall composition of an alloy. If the alloy does not dissociate into its constituents (show phase separation), there would be random lattice occupation or ordering in the arrangement. The degree of strain, growth conditions and thickness of the layer determine phase separation [5–7].

The 5UDH-2 Pelletron Tandem Accelerator ion irradiation has been often used to investigate the applicability of devices in irradiation environment. Previously, an investigation was conducted on protons, neutrons, neon and krypton ions irradiations to probe the radiation tolerance of InGaN thin film [8–11]. However, the irradiation energy was kept below 5 MeV. Very little is known about the influence of heavy ion irradiations above 5 MeV on InGaN material. In this material, phase separation into its constituents under irradiation environment is still a matter of debate. The phase separation and defects production in ion irradiated materials could have adverse effects on the usability of InGaN based devices in optical applications. This study could provide a platform for understanding the behavior of InGaN thin film material interaction with ion irradiation specifically with Si ion irradiation.

In this study, tailoring the structural and optical characteristics of InGaN/GaN multilayer thin film by 12 MeV silicon (Si) ions beam irradiations, are discussed. X-ray diffraction (XRD) and Raman shift

* Corresponding author. Tel.: +923339336761.
E-mail addresses: shakphy@gmail.com, shakilkhan_16@pieas.edu.pk (S. Khan), abdulwaheed32@gmail.com (A. Waheed).

http://dx.doi.org/10.1016/j.mssp.2017.03.004
Received 16 June 2016; Received in revised form 21 February 2017; Accepted 5 March 2017

investigations were employed to investigate the consequences of different fluences of Si ions irradiation on the structural modulations. Ion beam irradiation induced change in the optical bandgap was also studied.

2. Experimental details

InGaN/GaN thin film grown on sapphire substrate has been used in this research work. It was purchased from Xiamen Powerway Advanced Material Co., Ltd., China. As grown film was exposed to a collimated beam of Si ions with 12.0 MeV energy provided by a 5UDH-2 Pelletron Tandem Accelerator, installed at National Centre for Physics (NCP) in Islamabad, Pakistan. The vacuum was kept at 10^{-6} Torr during the irradiations process. The projected range of 12 MeV Si ions in the InGaN was found to be about 4.08 μm using the stopping/range of ions in matter code (SRIM 2008). The range is kept much larger than the film thickness for ensuring a homogenous damage distribution. The samples were mounted on a high-precision goniometer so that the orientation of samples relative to Si ion beam could be precisely controlled. The experiments were performed at different ion fluences at 1×10^{13}, 1×10^{14} and 1×10^{15} ions cm^{-2}. The fourth sample was not irradiated and had been taken as a reference sample.

For structural analysis of pristine and irradiated films, X-ray diffraction (XRD) and Raman spectroscopy was utilized. The XRD data were collected in the 2θ range from 20° to 80° with a step size of 0.02° using a Bruker X-ray diffractometer, operated at 40 kV, 40 mA and at a temperature of 25 °C, with Cu-K_α (λ = 1.54191 Å) radiation. Rutherford backscattering spectroscopy (RBS) was used to investigate quantitative and qualitative elemental composition. The Raman spectra were collected by a LabRAM III from DongWoo Optron, South Korea, using a 150 mW argon ion laser with 514 nm wavelength. For optical characteristics, UV–Vis spectrophotometer was utilized.

3. Results and discussion

3.1. X-ray diffraction analysis

XRD spectra shown in Fig. 1 of InGaN/GaN epilayer were taken before and after irradiation for obtaining information of irradiation induced structural changes. In as grown film spectrum, two peaks are observed at 33.92° and 34.74°. The prominent peak is related to (0 0 2) orientation of InGaN while relatively lower intensity peak correspond to (0 0 2) orientation of GaN. The broader and smaller peak for GaN observed in pristine spectrum is due to reflection from the underlying

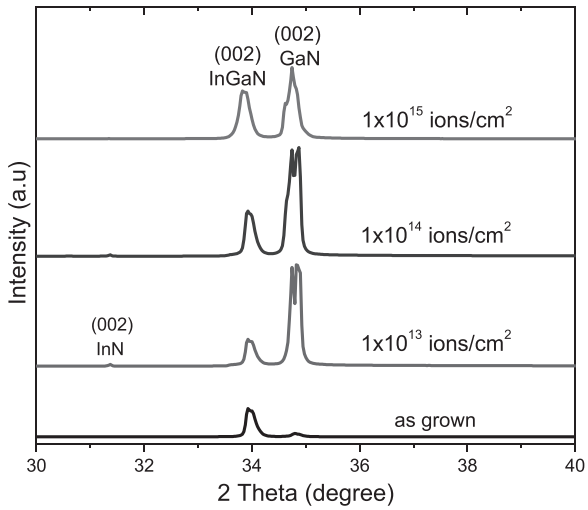

Fig. 1. XRD spectra of InGaN/GaN pristine and Si^+ irradiated epilyers at different fluences.

GaN layer. After ion irradiation at 1×10^{13} ions/cm^2, GaN peak intensity is abruptly enhanced compared to InGaN phase and another lower intensity peak appears at 31.36° corresponding to hexagonal (0 0 2) orientation of InN (Reference card:00-002-1450). However, with further increase of the fluence to 1×10^{15} ions/cm^2, the GaN peak intensity decreases relative to InGaN peak.

To clearly observe the above mentioned changes, the region of interest in XRD patterns has been enlarged and is shown in Fig. 2. Fig. 2(a), shows that the InN peak intensity increases at a fluence of 1×10^{13} ions/cm^2. However, when the fluence is increased to 1×10^{15} ions/cm^2, this peak broadens, indicating the amorphization of InN phase. The enlarged region of other two peaks of InGaN and GaN is shown in Fig. 2(b). The InGaN peak is slightly shifted after ions irradiation. At low fluence of 1×10^{13} ions/cm^2, the InGaN peak remains stable. With the increase of fluence, this peak shifts toward smaller angles and the shift at 1×10^{15} ions/cm^2 is slightly higher as compare to the shift occurs at 1×10^{14} ions/cm^2 ion dose. As far as the GaN peak is concerned, its intensity not only increases but also splits into two sub peaks (at 34.7° and 34.9°) at the ion fluence of 1×10^{13} ions/cm^2 and 1×10^{14} ions/cm^2. However, these two peaks are not evident at higher ion fluence of 1×10^{15} ions/cm^2.

Grain/particulate size of InGaN in pristine and irradiated film has been calculated using the Scherrer's formula $D = 0.9\lambda / B\cos\theta$ (D is the crystallite size, λ is incident radiation wavelength (Cu K_α = 0.154 nm) and B is FWHM) [12]. The crystallite sizes of InGaN before and after irradiation were calculated from using FWHM and are listed in Table 1. It can be seen that grain size of the InGaN phase is not much affected due to ion irradiation.

Appearance of InN and GaN peaks may correspond to ion beam induced nucleation of new grains or segregation of existing phase. The phenomena can be well understood from thermal spike (TS) model [13]. Ions in MeV energy range interact with InGaN/GaN and cause significant amount of electronic energy loss along their track. It also ionizes the InGaN/GaN along the track thereby transferring energy to its lattice and results in localized heat production along the ion track, which is known as thermal spike. The localized heating creates transient molten zones at several thousand of degree Kelvin which result in atomic displacement and mass movement in InGaN/GaN system [14]. In picoseconds time frame, ion tracks cool down (quenching) resulting in phase segregation and ultimately producing InN and GaN phases. Different phase clusters form after ion beam induced local heating and rapid quenching. It can also be observed from Fig. 1 that GaN relative peak intensity first increases with increasing Si^+ irradiation and then decreases at higher ion fluence (1×10^{15} ions cm^{-2}), possibly due to the generation of defects that consequently leads to lower degree of GaN phase crystallinity.

The deviations of thin film peak position from that of its corresponding bulk material value represent the stress development during or post growth treatment. The expansion of lattice parameters as evident from negative shift in InGaN peak positions of the irradiated films may be associated with the stress generation during film irradiation and demonstrate a tensile stress [12,15]. It may also be due to entrapment of Si atoms which act as impurities within the InGaN lattice [16]. The rise in peak intensity with increase of ion fluence could be related to local temperature increase as proposed in thermal spike model. The local rise in temperature (annealing) for a very short span of time increases the total lattice energy. This induces crystallization in the film that consequently leads to increase in peak intensities, grain sizes and reduction in defects [17].

As far as GaN peak splitting is concerned, it may be due to the formation of a damaged band, in GaN layer. Due to irradiation, in addition to the formation of surface damaged layer, the ion beam-induced-point-defects are produced and distinct lattice disorder along with lattice expansion occurs [18,19]. These phenomena may be the reason for splitting of peaks (at 2θ = 34.74° and 34.86°) observed for GaN. The disappearance of peak splitting at higher ion fluence may be