Influence of High Energy Radiation on the Structural Properties of Gallium Nitride Films

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Abstract: Gallium Nitride (GaN) thin films were fabricated via the molecular beam epitaxy (MBE) technique. A plasma source with a radio frequency (rf) plasma source was used --as the nitrogen source. A conventional effusion cell was used to provide Ga flux. The samples used in this study were grown on two substrates types: on Si <111> substrates and sapphire <0001> substrates. The used samples were exposed to three different gamma doses (γ) of 1600 kGy, 3700 kGy, 5300 kGy using Cobalt-60 source. The effect of gamma radiation on the structural properties of GaN films has been systematically studied via a computer-aided X-ray diffractometer. The results confirm that the first, second and third dose of gamma caused shifting in 20 towards the lower side, and that shifting increase with an increase in the radiation dose in the GaN film. The FWHM was widened and reduced the intensity ratio (IA/IB) by increasing the gamma radiation dose from 1600 kGy to 3700 kGy and 5300 kGy. The effect of the thin film thickness and the N/Ga ratio on the structural properties of "Film quality" was also investigated before and after γ -radiation. The results show a decrease in the film quality after exposing the film to films to γ -radiation. This decrease in film quality may be due to creating crystal defects and disorders in the crystal lattice. These defects and disorders increased with increasing the dose of γ -radiation.

Keywords: Gallium nitride, Radiation, Semiconductor materials, molecular beam epitaxy, metal organic chemical vapour deposition

1. Introduction

Gallium nitride (GaN) shows a several unique properties, such as large direct band gap, strong inter-atomic bonds, and high thermal conductivity, making it an ideal material for optoelectronic and high-power electronic devices [1]. More recently, high efficiency diodes based on GaN blue / green and blue lasers have been manufactured [2,3]. Despite the impressive advancements in GaN-made devices, the various defects roles in the material and their influence on the performance of the devices has not yet been understood, and thus it arouses great interest in investigating the behavior of deep defects, which can influence the structural, and optical properties of GaN thin films. On other hand, many researchers paid attention in investigation the effect of gamma and other ionizing radiation on the GaN devices due to increase the concerns related to the device's stability in environments of the radiation [4]. Several reports were discussed the defects induced in silicon Si [5,6], GaAs [7-9], InP [10,11] and ZnSe [12] by the radiation, but unfortunately a few reports were found to discuss the radiation influence on the structural properties of GaN thin films. In this work, we discussed the influence of gamma ray radiation on the structural properties of GaN thin film. Moreover, the effect of gamma rays on the thin film quality was discussed based on the defect's formation under gamma dose radiation.

2. Experimental

In this work the (GaN) thin films were fabricated via molecular beam epitaxy (MBE-Riber32P) technique equipped with a 2200 l/s turbo-molecular pump backed with a 50 CFM rotary roughing pump. A radio frequency (RF) plasma source was used as the nitrogen source. A conventional effusion cell was used to provide Ga flux. The samples used in this study were grown on two substrates types: on Si <111> substrates and sapphire <1000> substrates. The MBE system consists of three main vacuum chambers: a growth, a buffer, and a sample load lock chamber. After mounting the substrate by using a large carriage, it is outgassed in the preparation chamber to 140°C for 60 minutes to remove water vapor or other contaminants from the surface. After outgassing, the substrate is transferred to the growth chamber and is subjected to a nitridation process using the (RF) plasma source. The fabricated samples were exposed to three different doses of gamma (γ) radiation of values (1) =1600 kGy, (2) =3700 kGy, and (3) = 5300 kGy using Cobalt-60 source. The growth conditions for the used GaN films are summarized in Table 1.

Table 1: Growth	conditions	for the	used	GaN	films
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No. of Sample	Substrate Type	N/Ga Flu Ratio	Thickness (nm)
SA1	Al ₂ O ₃ (0001)	42	1260
SA2	Al ₂ O ₃ (0001)	42	1310
SA3	Al ₂ O ₃ (0001)	42	1020
SA4	Al ₂ O ₃ (0001)	24	1419
SI1	Si (111)	18	1274
SI2	Si (111)	20	1070
SI3	Si (111)	48.7	1030
SI4	Si (111)	34	604

3. Results and discussion

3.1 Effect of γ radiation on the structural Properties of GaN films grown on Sapphire <0001>

X-ray diffraction (XRD) is an extremely useful tool for the characterization of semiconductor heterostructures [13]. Important parameters of epitaxial layers such as absolute lattice constant, lattice mismatch with respect to the substrate, residual strain and alloy composition can be

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obtained with a high-resolution x-ray diffraction system [14]. In addition, film crystallinity can be qualitatively evaluated with such a tool. Such information is of critical importance for the optimization and characterization of the growth process. The influence of gamma radiation on the structural properties of GaN films has been examined by using X-ray. The Figure. 1, exhibit the XRD patterns of four GaN films grown on Al₂O₃(0001) Substrates before and after ydose radiation. Three different doses namely 1600, 3700, and 5300 kGy have been selected in this study. For better understanding and to make a clear idea about the effect of γ dose radiation on the GaN, we will discuss the effect of the three different doses of γ -radiation on the main diffraction GaN line which has (002) reflection. It is clear from these figures that there is a shift in the diffraction lines to lower 20 values after radiation. For (002) diffraction lines, the amount of 2θ shift was ranging from 0.02° after the first (lowest) γ -dose to 0.24° after the third (highest) γ dose. Also, the full width at half maximum (FWHM) of these lines is widened by an amount of 0.02° after the second dose to 0.06° after the third dose. In addition to the previous changes, there was a decrease in the intensity of these lines after radiation by an amount ranging from 14% after the first dose to 33.6% after the third dose. From these results it is clear that the process of decreasing Ga/N ratio with increasing the sample thickness is playing a clear role in the effect of radiation on the diffraction pattern of GaN films. This is clear from results of the first three samples (SA1, SA2, and SA3) that have the same Ga/N ratio (42%) and convergent thicknesses of 1260, 1310, and 1020 nm respectively. For these three samples there was a shift of 2θ of amount 0.24° from 34.51° before radiation to 34.27° after

the third y-radiation dose. For sample SA4 of larger thickness (1419 nm) and lower Ga/N ratio (24%), 20 is shifted by an amount of 0.22° from 34.51° before radiation to 34.29° after the third γ -radiation dose. That means that 20 shift is less for the thicker and lower Ga/N ratio films. For the four samples, it is obvious that the FWHM was widened 0.06° after the third dose of radiation. It is obvious from FWHM before radiation, that sample SA4 has the highest quality because it has the lowest FWHM value of 0.12°, which means that as the films go thicker its quality go better as reported by researchers [15], this may be due to increasing the film homogeneity, and decreasing the crystal defects with increasing the film thickness. From these Fig. 2 and Table 1, it is obvious that the first dose of γ radiation (1600 kGy) had a small effect on the structural properties where it didn't increase the FWHM, it caused only a small shift in 20 between 0.02° and 0.04° and a decrease in the diffraction line intensity by an amount ranging between 12% and 14%. Also, it is noticeable that the second dose (3700 kGy) and the third dose (5300 kGy) had a clear effect on the structural properties where 2θ was shifted more to lower values, the FWHM was widened, and the intensity ratio (I_A/I_B) was reduced strongly by increasing the γ radiation dose from 1600 kGy to 3700 kGy and to 5300 kGy. Also, from these results it is clear that the thickness and N/Ga ratio had a clear effect on the structural properties of GaN films, whereas the thickness increasing and Ga/N ratio decreasing the quality of the film is better and the effect of γ radiation is less (as shown in the figures and tables). Similar observation was reported by different researcher in different materials such as TeO2 Thin Films [16], and CuO [17].



Figure 1: XRD patterns before and after Gamma Radiation for the prepared GaN films grown on Sapphire Al₂O₃(0001)

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Table1: Overview on The effect of the three -radiation doses, [dose (1) =1600 kGy, dose (2) =3700 kGy, and dose (3) =5300 kGy], on the main line (002) of the diffraction patterns for the prepared GaN films grown on Sapphire $Al_2O_3(0001)$

Sample	SA1	SA2	SA3	SA4
N/Ga Flux ratio	42	42	42	24
Thickness (nm)	1260	1310	1020	1419
2θ (°) Before γ radiation	34.51	34.51	34.51	34.51
FWHM (°) Before γ dose	0.14	0.14	0.16	0.12
I_A/I_B (%) After γ dose (1)	86.9	86.9	86	88
I_A/I_B (%) After γ dose (2)	81	80.6	79.7	81.3
I_A/I_B (%) After γ dose (3)	67	67.19	66.4	67.7
2θ (°) After γ dose (1)	34.47	34.47	34.47	34.49
2θ (°) After γ dose (2)	34.41	34.41	34.41	34.43
2θ (°) After γ dose (3)	34.27	34.27	34.27	34.29
FWHM (°) After γ dose (1)	0.14	0.14	0.16	0.12
FWHM (°) After γ dose (2)	0.16	0.16	0.18	0.14
FWHM (°) After γ dose (3)	0.2	0.2	0.22	0.18



Figure 2: A graphical representation of the variation of (a) the 2θ , and (b) FWHM before and after Gamma Radiation for the prepared GaN films grown on Sapphire Al₂O₃(0001).

3.2. Effect of γ radiation on the structural Properties of GaN films on silicon <111>

The effect of γ radiation on GaN films grown on Si (111) is shown in Fig. 3. For better and clear comparison, we will discuss the effect of the three different doses of y-radiation on the main diffraction GaN line which has (002) reflection. For the first sample (SI1) of largest thickness (1274nm), 20 was shifted by an amount of 0.36° to a lower value from 34.57° before radiation to 34.21° after the third dose of radiation, and also the same shift was for SI2 sample of thickness 1070 nm. But for Sample SI3, 2θ was shifted by a larger value of amount 0.38° from 34.57° before radiation to 34.19° after the third dose of radiation. This large shift of 2θ in SI3 may be is due to the increase is N/Ga ratio (48.7%). Also, for these first three samples, the FWHM was increased by an amount of 0.08° after the third dose of γ radiation. For sample SI4 of thickness 604 nm $.2\theta$ was shifted more (0.48°) than the other three samples from 34.57° before radiation to 34.09° after the third dose. Also, the FWMH of this

sample (SI4) was larger, where it was widened by an amount of 0.18° from 0.16° before radiation to 0.34° after the third dose of radiation. Regarding the ratio of diffraction lines intensities after and before y-radiation (Iaftre/Ibefor), It was varied form 57.1% for the sample of larger thickness (1274 nm) to 47.6% for the sample of the smallest thickness (604 nm) after the third dose of radiation. From these results, the first dose of γ radiation (1600 kGy) had a small effect on the structural properties where it didn't increase the FWHM, It caused only a small shift in 2θ between 0.04° and 0.08° and a decrease in the diffraction line intensity by an amount ranging between 17% and 25%. Also, it is noticeable that the second dose (3700 kGy) and the third dose (5300 kGy) had a clear effect on the structural properties where 2θ was shifted more to lower values, the FWHM was widened, and the intensity ratio (I_A/I_B) was reduced strongly by increasing the γ radiation dose from 1600 kGy to 3700 kGy and to 5300 kGy. It is clear from all these results that the thickness and N/Ga ratio had a clear effect on the structural properties of GaN films, whereas the thickness is going large the quality

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of GaN films is better which agrees with the other researchers [48] (as explained in the previous section), whereas N/Ga ratio is going large the quality of the film is less. Also from these results, the effect of substrates type was obvious on the structural properties of GaN films before and after γ -radiation, where the films grown on AL₂O₃ (0001) had a better quality (before and after γ radiation) than

the films grown on si (111). Thismay be due to the matching between GaN films and the substrate type; where this matching is better in case of AL2O3(0001) substrates. We can conclude that: the quality of GaN films decreased after exposing the films to high doses of γ -radiation. This decrease in the film quality may be is due to creating of crystal defects and disorders in the crystal lattice.



Figure 3: XRD patterns before and after Gamma Radiation for the prepared GaN films grown on Si (111)

Table 1: Overview on The effect of the three -radiation doses, [dose (1) =1600 kGy, dose (2) =3700 kGy, and dose (3) =5300 kGy], on the main line (002) of the diffraction patterns for the prepared GaN films grown on Si (111).

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Sample	SA1	SA2	SA3	SA4
N/Ga Flux ratio	18	20	48.7	34
Thickness (nm)	1274	1070	1030	604
2θ (°) Before γ radiation	34.57	34.57	34.57	34.57
FWHM (°) Before γ dose	0.14	0.16	0.16	0.16
I_A/I_B (%) After γ dose (1)	83	82	81.5	75
I_A/I_B (%) After γ dose (2)	69.1	68	67	57.14
I_A/I_B (%) After γ dose (3)	57.1	56.6	55.8	47.6
2θ (°) After γ dose (1)	34.53	34.53	34.51	34.49
2θ (°) After γ dose (2)	34.37	34.37	34.35	34.25
2θ (°) After γ dose (3)	34.21	34.21	34.19	34.09
FWHM (°) After γ dose (1)	0.14	0.16	0.16	0.16
FWHM (°) After γ dose (2)	0.16	0.2	0.2	0.28
FWHM (°) After γ dose (3)	0.22	0.24	0.24	0.34

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Figure 2: A graphical representation of the variation of (a) the 2θ, and (b) FWHM before and after Gamma Radiation for the prepared GaN films grown on Si (111)

4. Conclusion

From the above work we can conclude that the first dose of γ radiation (1600 kGy) had a small effect on the structural properties of GaN films grown on Sapphire (0001) substrates. where it didn't increase the FWHM, it caused only a small shift in 20 between 0.02° and 0.04° and a decrease in the diffraction line intensity by an amount ranging between 12% and 14%. The second dose (3700 kGy) and the third dose (5300 kGy) had a clear effect on the structural properties where 2θ was shifted more to lower values, the FWHM was widened, and the intensity ratio (I_A/I_B) was reduced strongly by increasing the γ radiation dose from 1600 kGy to 3700 kGy and to 5300 kGy. The thickness and N/Ga ratio had a clear effect on the structural properties of GaN films, whereas the thickness increasing and Ga/N ratio decreasing the quality of the film was better and the effect of γ radiation is less. With respect to the films grown in Si(111), the first dose of γ radiation (1600 kGy) has a small effect on the structural properties GaN films, where it didn't increase the FWHM, It caused only a small shift in 20 between 0.04° and 0.08° and a decrease in the diffraction line intensity by an amount ranging between 17% and 25%. The second dose (3700 kGy) and the third dose (5300 kGy) had a clear effect on the structural properties where 2θ was shifted more to lower values, the FWHM was widened, and the intensity ratio (I_A/I_B) was reduced strongly by increasing the γ radiation dose from 1600 kGy to 3700 kGy and to 5300 kGy. The thickness and N/Ga ratio had a clear effect on the structural properties of GaN films, whereas the thickness is going large the quality of GaN films is better which agrees with the other researchers, whereas N/Ga ratio is going large the quality of the film is less. The effect of substrates type was obvious on the structural properties of GaN films before and after yradiation, where the films grown on AL₂O₃ (0001) had a better quality (before and after γ radiation) than the films grown onSi (111). Finally, we can conclude that: the quality of GaN films decreased after exposing the films to yradiation. This decrease in the film quality may be is due to creating of crystal defects and disorders in the crystal lattice. These defects and disorders increased with increasing the dose of γ -radiation.

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