The Influence of Gamma-Ray-Induced Deep Defects on the Luminescent Properties of Sputtered GaN Thin Films

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ABSTRACT

The structural and optical properties of rf magnetron-sputtered GaN thin films on p⁺-Si substrates have been accessed as a function of γ-ray irradiation doses from 1 to 12 Mrad. Analytical results revealed that the increasing dose of γ-rays could enhance the more occurrence of nitrogen vacancies which not only created a prominent deep level luminescence but also destroyed the crystallinity of GaN thin films. For low dose of γ-ray irradiation (≤ 4 Mrad (GaN)), evidence showed that by raising the irradiation dose, more associated Ga-H complexes will be effectively promoted, yielding an enhanced yellow band emission. However, for high dose of γ-ray irradiation (> 4 Mrad (GaN)), further higher dose of γ-rays could lead the dissociation of Ga-H complexes in GaN samples, resulting in a repressed yellow band emission.

(KEYWORDS: GaN; γ-ray; nitrogen vacancies; crystallinity; Ga-H complexes; yellow band emission)
I. INTRODUCTION

Gallium nitride (GaN) is a chemically stable compound semiconductor with a wide direct band gap (3.4 eV). This makes GaN attractive for application of UV/blue light emitting diodes and high-power and –temperature devices. GaN-based electronic and optoelectronic devices, including visible light emitting diodes (LED), metal-semiconductor field-effect transistor (MESFET), high electron mobility transistor (HEMT), UV photoconductive detector, UV photovoltaic detectors, have been fabricated [1-2]. Despite the impressive progress in GaN-made devices, the role of various defects in the material and their effect on device performance are not yet understood and therefore cause intense interest in studying the behavior of deep defects which affects the microstructural and luminescent properties of GaN thin film.

Among the defect-induced transitions, the ubiquitous yellow luminescence (YL) [3-4] observed from photoluminescence (PL) at around 2.2 ~ 2.3 eV, is detected independently of the substrate and the epitaxial technique used, with its intensity being more pronounced at room temperature. Although the YL measurements reveal a distinct optical signature of the defect, the microstructure and chemical nature of the defects has not been identified.

On the other hand, there is also a growing interest in the effect of gamma and other ionizing radiation on GaN devices because of the increasing concerns related to the stability of devices in radiation environments. Many papers have reported radiation-induced defects in silicon [6], GaAs [7-8], and ZnSe [9] materials, but up to now little work has been done to connect irradiation effects on the microstructural and luminescent properties.

In this article, the effects of γ-ray irradiation on the defect status of radio-frequency-sputtered GaN thin films are systematically investigated. Moreover, in addition to the variation of γ-ray-irradiated defects, the results of microstructural and luminescent characteristics of GaN thin films in relation to the dose of γ-ray irradiation are also understood.

II. EXPERIMENTAL
GaN thin films were prepared by a two-stage-growth [10] of radio-frequency (rf) magnetron sputtering method. The source material, made by Superconductive Components, Inc. (USA), was GaN target (3 inch diam.) with 99.999 % purity. Prior to loading the p⁺-Si (111) substrates into the sputter chamber, the wafers were rinsed in 10:1 HF acid to remove the native oxide layer and blown dry. During the growth process, the rf power was controlled at 50W and a mixed sputtering gas of N₂ and Ar was utilized to keep the sputtered pressure at $5 \times 10^{-3}$ Torr. For the first stage, a thin GaN buffer layer ($\approx 500$ Å ) was deposited on Si substrate at 400°C. Then, the substrate temperature was elevated to 500°C for annealing the GaN buffer under a purified N₂ atmosphere for 30 min. Next, a 3500Å thickness of GaN epilayer was grown on top of GaN buffer at 700°C. After deposition, rapid thermal annealing (RTA) treatment at 900°C on the sputtered GaN thin films were carried out in 0.5 Torr purified N₂ ambient for 1 min. Subsequently, some RTA-treated GaN thin films were arranged to expose to the γ-ray irradiation of Co⁶⁰ with various doses (1 ~ 12 Mrad (GaN)) under a dosing rate of 100 rads/sec.

The crystallinity of GaN thin films was examined using a x-ray diffraction (XRD) with a Cu-Kα radiation source. Luminescent analysis of different doses of γ-ray-irradiated samples were carried out employing PL measurements excited by a 325 nm He-Cd laser at room temperature. Compositional analyses of different doses of γ-ray-irradiated samples and their comparisons with pre-irradiated ones were performed employing secondary ion mass spectrometry (SIMS). The accuracy of the SIMS data in depth of film thickness and in concentration of tested elements are within 5% and with 3% relative error, respectively. Fourier Transform Infrared Spectroscopy (FTIR) measurements were carried out using a MAGNA-IR 750 SPECTROMETER. The resolution of the spectrometer was 4 cm⁻¹, and all spectra given are expressed in absorbance. For the deep level transient spectroscopy (DLTS) measurement [11], Schottky diodes were made by evaporating gold (Au) and aluminum (Al) on top of GaN thin film which provide, respectively, a Schottky contact at Au/GaN and an Ohmic contact at Al/GaN junctions.

**III. RESULTS AND DISCUSSION**

The GaN thin films pre-irradiated and γ-ray irradiated were all found to be $n$-type from
room temperature Hall-effect measurements, as is usual case for undoped GaN. Typical XRD profiles of GaN thin films pre-irradiated and γ-ray irradiated at various doses are shown in Fig. 1, indicating that the crystallinity was strongly influenced by the γ-ray irradiation. As illustrated in Fig. 1, the intensity of dominant peak in pre-irradiated GaN located at $2\theta = 32.4^\circ$, originating from (1010) wurtzite ($\alpha$)-GaN, evidently decreases with the increasing doses of γ-ray irradiation. It implies that most of irradiation energy might take on the destructive role deteriorating the crystallinity of GaN film.

In general, it is expected that for thin films with superior crystallized property, a well-performing band-to-band transition in photoluminescence should result. Such an expectation was realized by our photoluminescence of different doses of γ-ray-irradiated GaN thin films. As illustrated in Fig. 2, the strong near-band-edge emission at 3.3 eV for pre-irradiated sample was observed to decrease with the increasing dose. This band emission has been assigned to excitons bound to neutral donors [12] and is considered a PL fingerprint of N-type GaN. It is worth noting that a significant phenomenon illustrating the intensity of a weak deep level luminescence at 2.8 eV for pre-irradiated specimen contrarily increased with increasing irradiation dose. Such a result implies that the deep level luminescence not only affects near-band-edge emission but also plays the destructive role in determining the crystallinity of GaN thin film. Another interesting observance showing the intensity of a broad yellow band around 2.2 eV for pre-irradiated sample increases with the increase of irradiation dose for low dose of γ-ray irradiation ($\leq 4$ Mrad (GaN)). However, for high dose of γ-ray irradiation ($> 4$ Mrad (GaN)), the intensity of yellow band emission contrarily decreases with the increasing irradiation dose. It implies that low dose ($\leq 4$ Mrad (GaN)) and high dose ($> 4$ Mrad (GaN)) of γ-ray irradiation play the promotive and repressive roles, respectively, in yellow band emission of GaN samples.

To investigate the physical origins for the variation of PL which occurred in our samples, DLTS measurements were employed to examine the deep defects existing within these GaN thin films. The DLTS results are shown in Fig. 3. As can be seen, two deep electron traps (designated as $E_{t1}$ and $E_{t2}$, respectively) were consistently detected in all the samples. Moreover, the Arrhenius plots yielding the activation energies of $E_{t1}$ and $E_{t2}$ below the conduction band-edge minimum are $E_c - E_{t1} \approx 0.59$ eV ($\pm 0.02$ eV) and $E_c - E_{t2} \approx 0.82$ eV ($\pm$
0.02 eV). First, we suspect that E_{t1} is a nitrogen-vacancy-related deep trap. This inference is based on the SIMS observation that the higher dose of γ-ray-irradiated sample the more nitrogen deficiency detected inside the GaN bulk, as illustrated in Fig. 4. Moreover, the measured E_{t1} level at 0.59 eV below the conduction band in this experiment is quite in agreement with the nitrogen-vacancy-related deep level at 0.62 eV which was observed by thermally stimulated current (TSC) method [13]. Significantly, the observance of the DLTS signal intensity of E_{t1} in pre-irradiated sample enhanced by raising the dose of γ-ray irradiation provides a reliable deduction that E_{t1} not only is the causality of deep level luminescence at 2.8 eV but also behaves the main factor in destroying the crystallinity of GaN film. Next, the origin of E_{t2} is suggested to be a hydrogen-related deep electron trap since the FTIR results revealed such an assertion. In addition, the measured E_{t2} level at 0.82 eV below the conduction band in this work is also coincident with report by Pearton et al. [14], that a hydrogen-induced electron trap at around E_c − 0.8 eV was found for implantation of hydrogen into GaN. Typical FTIR absorption spectra from the GaN samples are shown in Fig. 5, in which the spectral region is $1500 \sim 2000$ cm\(^{-1}\). There is no observable absorption band except background from the pre-irradiated GaN samples, as shown by the curve (a) in Fig. 5. After the samples were irradiated by low dose of γ-rays ($\leq 4$ Mrad (GaN)), one absorption band around 1730 cm\(^{-1}\) was observed to be enhanced and reached a maximum by raising the dose of γ-rays until 4 Mrad (GaN) (see curves (b) and (c) of Fig. 5). Contrarily, for the case of high dose of γ-rays (> 4 Mrad (GaN)), increasing the irradiation dose of γ-rays significantly causes the intensities of the above absorption bands to decrease as shown by curves (d) and (e) of Fig. 5. The origin of FTIR band at around 1730 cm\(^{-1}\) has been ascribed as the local vibrational modes of Ga-H complexes in the vicinity of N vacancies [15].

Based on the above results, we have the following deduction. Before the γ-ray irradiation, hydrogen could be situated in either IR inactivated states or very weak IR activated states. For the low dose of γ-ray irradiation ($\leq 4$ Mrad (GaN)), γ-ray irradiation can sufficiently dissociate hydrogen-related complexes in IR inactivated states and result in atomic hydrogen. Atomic hydrogen is very active and can combine with Ga dangling bonds beside the N vacancies to form Ga-H complex. However, as in the high dose of γ-ray irradiation (> 4 Mrad (GaN)), further higher dose of γ-rays not only introduces more defects but also lead to the dissociation of H complexes containing Ga-H complexes in GaN samples.
IV. CONCLUSIONS

We have performed different doses of $\gamma$-ray irradiation treatment on undoped GaN samples. Two deep electron traps, $E_{t1}$ and $E_{t2}$ ($E_c - E_{t1} \approx 0.59$ eV and $E_c - E_{t2} \approx 0.82$ eV), corresponding $V_N$-related defect and Ga-H complex, respectively, were demonstrated to be strongly influenced by different doses of $\gamma$-ray irradiation. The $E_{t1}$ trap was observed to be enhanced by raising the irradiation dose and was deduced to be the main causality yielding the poor crystallinity as well as a deteriorated near-band-edge emission of GaN thin film. Interestingly, the $E_{t2}$ trap increasing with the increases of $\gamma$-rays at low dose of irradiation ($\leq 4$ Mrad (GaN)) and decreasing with the increases of $\gamma$-rays at high dose of irradiation ($> 4$ Mrad (GaN)) was suggested to play the prominent role in influencing the YB luminescent behavior.

V. ACKNOWLEDGMENT

One of the authors (Ching-Wu Wang) gratefully acknowledges the financial support from the National Science Council (NSC) in Taiwan under contract number: NSC89-2215-E-214-004.

REFERENCES

Fig. 1. X-ray diffraction spectra for sputtered-GaN thin films with different doses of γ-ray irradiation. Curve (a) represents the pre-irradiated sample. Curves (b) – (e) correspond to samples treated by 1, 4, 8, and 12 Mrad (GaN) of γ-ray irradiation.
Fig. 2. Room-temperature photoluminescence spectra for sputtered GaN thin films treated by different doses of γ-ray irradiation. Curve (a) represents the pre-irradiated sample. Curves (b) – (e) correspond samples treated by 1, 4, 8, and 12 Mrad (GaN) of γ-ray irradiation.
Fig. 3. DLTS signals of different doses of γ-ray-irradiated Au/GaN Schottky diodes. Curve (a) represents the pre-irradiated sample. Curves (b)–(e) correspond to samples treated by 1, 4, 8, and 12 Mrad (GaN) of γ-ray irradiation.
Fig. 4. SIMS depth profiles of N on GaN (epilayer)/GaN (buffer)/p'-Si devices with different doses of γ-ray irradiation. Curve (a) represents the pre-irradiated sample. Curves (b) – (e) correspond to samples treated by 1, 4, 8, and 12 Mrad (GaN) of γ-ray irradiation.
Fig. 5. FTIR spectra for sputtered-GaN thin films with different doses of γ-ray irradiation. Curve (a) represents the pre-irradiated sample. Curves (b) – (e) correspond samples treated by 1, 4, 8, and 12 Mrad (GaN) of γ-ray irradiation.