Radiation Effects on InGaN Quantum Wells and GaN Simultaneously Probed by Ion Beam-Induced Luminescence

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Abstract—InGaN quantum well structures on GaN epilayers were exposed to 500 keV alpha particles to fluences above $10^{14}$ cm$^{-2}$ to probe the relative radiation tolerance of the epilayer and wells. Performance was estimated by the intensity of ion-beam induced luminescence. Two separate types of quantum well structures emitted at 470 and 510 nm prior to irradiation, and only small wavelength shifts were observed even with the highest alpha fluences. Complementary cathodoluminescence experiments showed that luminescence in the quantum wells is strongly influenced by charges injected deep into the GaN epilayer. The 500 keV alpha penetration depth was $\sim 1 \mu$m, so that defects were created at a faster rate in GaN compared to InGaN as alpha particles slowed and stopped within a minority carrier diffusion length of the quantum wells. However, the rate of luminescent decay was similar for both materials. Taken together with the cathodoluminescence data, this ion-beam induced luminescence comparison indicates that the quantum well luminescence decay rate is dominated by radiation-induced defects in the GaN epilayer. InGaN quantum wells are then demonstrated to be not more than a factor of ten more radiation sensitive than GaN, and may be substantially less sensitive than this upper bound.

Index Terms—Alpha particle, InGaN, light-emitting diode, quantum well, radiation.

I. INTRODUCTION

InGaN quantum well structures can be used as the active region of efficient light-emitting diodes, especially in the wavelength range spanning 400–520 nm, where applications including optical communication systems and components in white light sources are being considered[1]. However, to allow operation of these devices in space and other radiation-rich environments, it is important to understand and control the effects of radiation-induced defects. Previous studies have employed penetrating particles such as neutrons and high-energy protons to probe the radiation tolerance of InGaN light-emitting diodes (LEDs)[1] and GaN double-heterojunction light-emitting diodes (LEDs)[2], but these experiments have necessarily modified not only the quantum well structures but also the thick GaN epilayer immediately underneath. The measured performance limitations therefore reflect the combined radiation tolerance of the composite system without providing a direct measurement of the light-generating InGaN quantum well layers. Wu et al. have reported high resistance to radiation damage from 2 MeV protons in In$_{1-x}$Ga$_x$N thin film alloys grown on sapphire substrates, which suggests that devices made with this material may have similar advantageous properties[3].

In this work, we perform in-situ measurement of ion-beam induced luminescence to simultaneously observe the properties of a five-layer stack of InGaN quantum wells and its GaN epilayer when these materials are irradiated with 500 keV alpha particles. We spectroscopically separate emissions from quantum wells and GaN, enabling a more independent estimation of radiation tolerance of the two structures. Next, we perform complementary electron-beam experiments and modeling to quantitatively determine the recombination properties of excited electrons as a function of particle penetration depth. Finally, we model our in-situ luminescence results using Monte Carlo simulations of alpha induced lattice defect creation coupled with finite element drift-diffusion simulations.

II. EXPERIMENTAL DETAILS

Quantum wells on GaN epilayers were grown by metalorganic chemical vapor deposition on sapphire substrates. Our tested structures consisted of five bilayer periods of 10 nm GaN and 3 nm of In$_{x}$Ga$_{1-x}$N ($x$ between 0.1 and 0.2), on top of a $\sim 4 \mu$m thick layer of GaN. Two separate microstructures were examined. In the first, with In$_{x}$Ga$_{1-x}$N, $x \approx 0.1$, there was a high density of lateral inhomogeneities such as V-defects, as determined by transmission electron microscopy and atomic force microscopy [4], [5]. The primary photoluminescence peak was at 470 nm. In the second sample, with In$_{x}$Ga$_{1-x}$N, $x \approx 0.2$, smooth and homogeneous quantum well structures were concluded from well resolved X-ray diffraction patterns and very low growth surface morphology as identified in atomic force microscopy. For this sample, the primary photoluminescence peak was at 510 nm.

Ion beam-induced luminescence experiments were performed at the Lawrence Livermore National Laboratory (LLNL) 4 MeV Ion Beam Accelerator. Samples were mounted on a water-cooled aluminum plate at 45 degrees to the incoming diodes (LEDs)[1] and GaN double-heterojunction light-emitting diodes (LEDs)[2], but these experiments have necessarily modified not only the quantum well structures but also the thick GaN epilayer immediately underneath. The measured performance limitations therefore reflect the combined radiation tolerance of the composite system without providing a direct measurement of the light-generating InGaN quantum well layers. Wu et al. have reported high resistance to radiation damage from 2 MeV protons in In$_{1-x}$Ga$_x$N thin film alloys grown on sapphire substrates, which suggests that devices made with this material may have similar advantageous properties[3].

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beam of 500 keV alpha particles, which was incident on the ~4 cm² samples at a current of 13 nA over a rectangular area of 14 mm by 3 mm. The resulting energy deposition rate, about 15 mW/cm², is not sufficient to generate a significant temperature increase in the quantum wells or epilayer during irradiation. Light was captured by a 5 mm diameter collimating lens positioned about 1 cm away from the sample, and transported via optical fiber to a spectrometer. In complementary electron beam experiments, the same light collection and spectral analysis system was employed, but light was generated by a rastered electron beam where current varied from 7 to 37 nA, with electron energies between 1 and 30 keV.

III. RESULTS

Fig. 1(a) shows several ion-beam induced luminescence spectra for the InₐGa₀.₉N, defect-rich quantum well structure, obtained sequentially as the sample was irradiated. The spectrum for the more homogeneous In₀.₂Ga₀.₈N sample, Fig. 1(b), is qualitatively similar. However, some differences, such as the rate of luminescence decay, are discussed in detail below.

A. Diffusion Length and Recombination

Fig. 1 demonstrates that the InGaN quantum well region emission is many times more intense when compared to emission from the GaN bulk, independent of fluence and sample microstructure. Because the total thickness of the quantum well layers is just 65 nm, it is unlikely that the quantum well signal is due to re-absorption processes in GaN. These keV electrons do not have sufficient energy to cause significant damage in the sample even after many minutes of continuous irradiation. The inset shows the ratio of the quantum well and GaN peak intensities as a function of electron beam energy. There is a small blue shift in the peak wavelengths with increasing electron energy. The spectrum for the more homogeneous In₀.₂Ga₀.₈N sample (data not shown) is qualitatively similar.

IV. DISCUSSION

Fig. 2 shows the electron beam-induced luminescence spectra from the same sample shown in Fig. 1(a). In contrast to the single alpha particle energy used in the ion-beam experiments, the electron energy was varied from 1 to 30 keV to provide insight into the relative radiative recombination efficiencies as a function of electron depth for the quantum well structure and underlying GaN. These keV electrons do not have sufficient energy to cause significant damage in the sample even after many minutes of continuous irradiation. The inset shows the ratio of the quantum well and GaN peak intensities as a function of electron beam energy. There is a small blue shift in the peak wavelengths with increasing electron energy. The spectrum for the more homogeneous In₀.₂Ga₀.₈N sample (data not shown) is qualitatively similar.
Fig. 3. Calculated cathodoluminescence intensity as a function of penetration depth of the electron beam for GaN/Ga0.45In0.55N quantum wells. The quantum well intensity rises rapidly and saturates, while the GaN signal grows steadily (drawn lines are guides to the eye).

[7]. This broad defect-related peak appears in the electron beam-induced spectra more prominently with increasing electron energy (and therefore penetration depth), indicating that it is indeed associated with defects in the underlying GaN layer, and not the quantum wells or with alpha-induced defects. Note also the slight blue shift which occurs for all the peaks with increasing fluence, which is possibly attributable to sample charging.

The relationship between emission and charge injection depth is more quantitatively demonstrated by results from electron-beam induced emission experiments (Fig. 2), and by the electron penetration depth calculations shown in Fig. 3. Here penetration depth is taken as the location where an increasing acceleration voltage results in the highest differential energy deposition. At 20 keV, which corresponds to a ∼ 3 : 2 ratio of quantum well to GaN emission signal, the electron penetration depth is about 710 nm—more than 10 times the total thickness of the quantum well stack.

B. Radiation Tolerance

The depth distribution of generated vacancy defects and deposited ionization energy was simulated using the SRIM-2006 code [8]. Fig. 4(a) shows the vacancy profile for 500 keV alpha particles incident at 45 degrees on to the GaN surface. The deposited ionization energy provides the carrier excitation which subsequently is observed by radiative recombination. We assume a linear scaling of the rate of radiative recombination and ionization rate. With this assumption the simulation can be compared to the luminescence profile. Fig. 4(b) gives the energy band diagram of the piezoelectric quantum well structure along with the ionization rate as a function of position.

Fig. 4(a) and (b) also shows schematically the quantum well structure in the same length scale superimposed in the calculated ionization and vacancy profiles. This demonstrates that most of the ionization energy (Fig. 4(b), circles) from the 500 keV alpha particles is deposited in the underlying GaN, and that vacancies, which are associated with optical performance loss (Fig. 4(a), triangles), are predominantly generated there as well. This vacancy profile enables a direct comparison to experiments performed on similar samples using, for example, heavy ions [9] or protons [1]. It is expected that any particles which produce similar vacancy densities in these quantum well structures will produce similar effects in their luminescent properties due to the electrically active nature of these defects.

As shown in Fig. 4(a), the concentration of Ga and N vacancies were calculated as a function of alpha particle fluence from SRIM [8]. From Goodman et al. [10], these vacancies are the primary electrically active defects generated when GaN is exposed to energetic alpha particles. The N vacancy is an electron donor with energy 0.2 eV below the conduction band of GaN, while the Ga vacancy is an electron acceptor with energy 0.95 eV below the conduction band [10]. The capture cross section of these defects is not well-established, but values near $3 \times 10^{-15}$ cm$^2$ have been reported [11].

Using the spatial distribution of Ga and N vacancies estimated from SRIM and the energies of these defects taken from [10], we calculated the radiative and non-radiative recombination rates of electron-hole pairs using the Silvaco Atlas computer code [12]. Non-radiative recombination is modeled with trap-assisted recombination and with Shockley–Read–Hall (SRH) recombination, assuming the vacancies act as traps with the appropriate
energies. For any point within the quantum well/GaN structure, time dependent recombination rates were calculated as a function of alpha fluorescence, as shown in Fig. 5. Finally, by spatial and temporal integration, the total radiative recombination in the quantum well/GaN structure was calculated and directly compared to the spectroscopically measured values obtained as a function of fluence. The capture cross section of the traps could be varied to fit the experimental data. This comparison is shown in Fig. 6.

In Fig. 6, the measured quantum well and GaN contributions were taken from the peak values of alpha-luminescence spectra, shown in Fig. 1. The measured data for both quantum well samples are consistent with a trap cross section with value $\sim 1 \times 10^{-15} \text{ cm}^2$, which agrees relatively well with published values [13]. The rate of decay for the normalized radiative recombination as a function of fluence in the In$_{0.2}$Ga$_{0.8}$N sample is faster than for the In$_{0.1}$Ga$_{0.9}$N sample. This difference may result from the fact that the In$_{0.2}$Ga$_{0.8}$N had fewer defects prior to irradiation, or it may be related to the higher concentration of In. The limited size of our samples does not allow a clarification of this point. Even for homogeneous In$_{1-x}$Ga$_x$N layers, Wu et al. observe variability in the degree of radiation tolerance as a function of In concentration. The radiation tolerance does not simply scale with the In composition [3].

As expected from SRIM, the model confirms the faster degradation rate of GaN vs. that of the quantum wells due to the larger number of defects introduced in GaN at the end-of-range of the alpha particles. It is important to note that under implantation of Au ions at $\sim 400$ keV GaN is significantly less likely to be amorphized than In$_{0.1}$Ga$_{0.9}$N. This follows from Rutherford backscattering experiments by Wendler et al. [9] Therefore, the relative radiation tolerance of In$_{1-x}$Ga$_x$N quantum wells observed here (Fig. 6, solid vs. dashed lines) may be in spite of a larger number of defects than the SRIM data would suggest. The Rutherford backscattering result is surprising given the high radiation tolerance of In$_{1-x}$Ga$_x$N alloys reported by Wu et al. [3], and may indicate that it will be challenging to fabricate radiation-hard solar cells from these materials that contain both n- and p-type layers. For the quantum well devices reported in this work, p-type In$_{1-x}$Ga$_x$N layers are not employed, and we place only an upper bound on the radiation tolerance of these layered structures. This limit does not immediately imply the devices, as opposed to the n-type material examined by Wu et al. [3], are more intrinsically radiation tolerant than GaN. As demonstrated by the much higher efficiency of the quantum wells for radiative recombination compared to the homogeneous GaN epilayer (cf., Fig. 1, 470/510 nm peaks vs. 365 nm peak), quantum wells have different luminescent properties relative to homogeneous materials such as the layers examined by Wu et al., and may exhibit differences in radiation tolerance as well.

In previous studies of quantum-well base devices [1], [2], more penetrating particles were used to create defects in light-emitting diodes. For example 40 MeV protons, with a range of approximately 50 $\mu$m in GaN, were used to probe InGaN LEDs [1]. In this case, the rate of vacancy creation as a function of depth was constant in the top, electrically active, 3–5 $\mu$m thick layer of the GaInN/GaN stack. The observed performance degradation rate could be dominated either by the GaN or by the quantum wells, but the data did not allow independent determination of the relative radiation tolerance of the two materials.

This work, by contrast, demonstrates that the radiation-induced LED performance decay may have been influenced significantly by damage in the GaN epilayers. In our experiments, the spectroscopically-separated epilayer and quantum well emissions enable us to estimate the relative performance of the quantum wells vs. GaN. For example, Fig. 4(a) (right axis) shows the alpha particle-induced damage rates quantitatively: in GaN, vacancies are created at an average rate of $\sim 0.01$ vacancies/$\text{A ion}$, while in the quantum wells, vacancies are created ten times slower - a rate closer to 0.001 vacancies/$\text{A ion}$. However, in Fig. 6, we see that the luminescence decay rates for GaN and InGaN quantum wells are similar. This demonstrates that the quantum wells are not worse by more than a factor of 10 compared to GaN. Otherwise, this would be reflected in a larger quantum well luminescence decay rate.
V. CONCLUSION

InGaN quantum wells and GaN epilayers were probed with 500 keV alpha particles and 1–30 keV electrons to determine their relative radiation tolerance via luminescence. The Monte Carlo model SRIM indicates ten times fewer defects were created in the quantum wells, compared to the GaN. However, radiation-induced luminescence decay rates were similar in both materials. Further, the relative alpha- and electron-induced luminescence in the InGaN and GaN demonstrates that most charges injected in the GaN recombined radiatively in quantum wells. Taken together, these observations mean that the luminescence decay rate in the quantum wells was dominated by defects generated in the GaN epilayer. The radiation sensitivity of InGaN quantum wells under our experimental conditions is therefore not greater than a factor of 10 compared to GaN, and it could be substantially smaller.

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REFERENCES


