COMPARISON OF THE OPTICAL PROPERTIES OF Er^{3+} DOPED GALLIUM NITRIDE PREPARED BY METALORGANIC MOLECULAR BEAM EPITAXY (MOMBE) AND SOLID SOURCE MOLECULAR BEAM EPITAXY (SSMBE)

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ABSTRACT

We report on the luminescence properties of Er doped GaN grown prepared by metalorganic molecular beam epitaxy (MOMBE) and solid-source molecular beam epitaxy (SSMBE) on Si substrates. Both types of samples emitted characteristic 1.54 µm PL resulting from the intra-4f Er^{3+} transition $^4I_{13/2} \rightarrow ^4I_{15/2}$. Under below-gap excitation the samples exhibited very similar 1.54 µm PL intensities. On the contrary, under above-gap excitation GaN: Er (SSMBE) showed ~80 times more intense 1.54 µm PL than GaN: Er (MOMBE). In addition, GaN: Er (SSMBE) also emitted intense green luminescence at 537 nm and 558 nm, which was not observed from GaN: Er (MOMBE). The average lifetime of the green PL was determined to be 10.8 µs at 15 K and 5.5 µs at room temperature. A preliminary lifetime analysis suggests that the decrease in lifetime is mainly due to the strong thermalization between the $^2H_{11/2}$ and $^4S_{3/2}$ excited states. Nonradiative decay processes are expected to only weakly affect the green luminescence.

INTRODUCTION

The luminescence from rare earth doped III-nitrides is of significant current interest for potential applications in optical communications and full color displays.[1] Visible and infrared electroluminescence (EL) has been reported from a number of rare earth doped GaN systems: GaN: Er (green, IR)[2,3,4], GaN: Pr (red, IR) [5], GaN: Eu (red) [6,7] and GaN: Tm (blue) [1] The incorporation, optical activation, and luminescence efficiency of rare earth ions in III-nitrides, however, is not yet fully understood. In this paper, we present a comparison of the PL properties of Er doped GaN grown by MOMBE and SSMBE. Excitation wavelength and temperature dependent PL studies were performed and analyzed in view of optoelectronic applications of Er doped GaN.

EXPERIMENTAL PROCEDURES

The GaN: Er (MOMBE) sample was grown in an INTEVAC Gas Source Gen II on In-mounted (100) Si substrate as described in reference [8]. The GaN: Er (SSMBE)

sample was grown on Si by solid source and RF-assisted molecular beam epitaxy (MBE). Details of the Riber MBE32 system used for growth have been discussed previously [3]. PL studies were performed using a HeCd laser operating at either 325 nm or 442 nm. Infrared PL spectra were recorded using a 1-m monochromator equipped with a liquid-nitrogen cooled Ge detector. In visible PL studies a thermo-electric cooled PMT was employed for detection. The signal was processed using lock-in techniques. IR lifetime studies employed the 355 nm line of a Nd: YAG laser for excitation. Visible lifetime data were taken by pumping into the $^4F_{7/2}$ Er^{3+} transition at ~495 nm.

RESULTS AND DISCUSSION

The infrared luminescence spectra of Er doped GaN grown by MOMBE and SSMBE are shown in Figure 1 for above (λ_{ex}=325 nm) and below-gap excitation (λ_{ex}=442 nm). The spectra were taken under identical experimental conditions. The pump power was kept constant at ~0.64 W/cm^2.
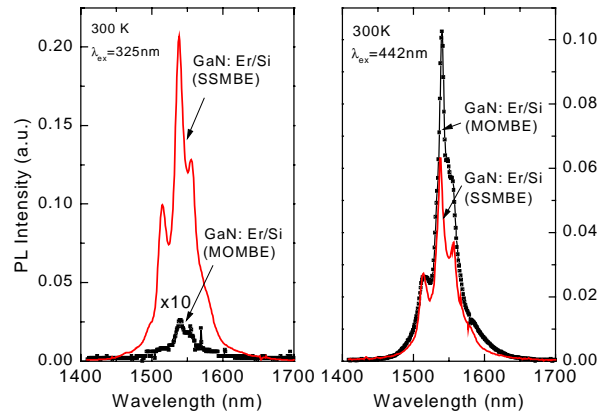

Figure 1: 1.54 μm PL spectra of GaN: Er (MOMBE) and GaN: Er (SSMBE) at room temperature. The PL was excited with either the 325 nm (above-gap) or 442 nm (below-gap) line of a HeCd laser.

Both samples exhibited characteristic 1.54 μm PL resulting from the intra-4f Er^{3+} transition $^4I_{13/2}\rightarrow^4I_{15/2}$. The most striking feature of figure 1 is the large difference in PL intensity observed for the samples under above-gap excitation. The GaN: Er (SSMBE) sample exhibited a strong 1.54 μm PL which was nearly 80 times more intense than that observed from GaN: Er (MOMBE). On the contrary, under below-gap excitation both samples exhibited very similar 1.54 μm PL intensities. As shown previously [9], the weak PL observed under above-gap excitation from GaN: Er (MOMBE) can be explained by a significantly reduced excitation efficiency compared to below-gap excitation. Visible PL studies (see below) revealed, that for GaN: Er (MOMBE) the bandedge provides an efficient radiative combination channel reducing the excitation efficiency of intra-4f Er

transitions. The excitation wavelength dependent PL study suggests that only weak electroluminescence can be expected from forward-biased GaN: Er (MOMBE) LED's.
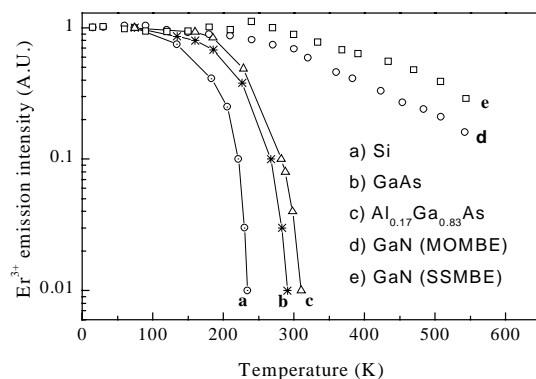

Figure 2: Comparison of the temperature dependence of the integrated Er^{3+} 1.54 µm PL for Er doped Si, GaAs, AlGaAs, and GaN (see also reference 10).

To further evaluate the GaN:Er samples for device applications, the temperature dependence of the integrated 1.54 µm PL intensities was measured as shown in Figure 2. Compared to Si: Er, GaAs: Er, and AlGaAs: Er [10], both GaN: Er samples exhibited very stable 1.54 µm PL up to temperatures as high as ~550 K. More information on the Er PL efficiency was obtained from temperature dependent lifetime studies. The total transition probability, i.e. the reciprocal of the experimental PL lifetime, is given as the sum of total radative decay rate, nonradiative decay rate through multiphonon relaxation, and nonradiative decay rate through energy transfer processes. It is assumed that at low temperature the nonradiative decay through either multiphonon relaxation and/or energy transfer is negligible small. Therefore, the lifetime at 15 K yields a good approximation for the radiative decay rate. Furthermore, assuming the radiative decay rate is temperature independent, any reduction in lifetime can be assigned to the onset of nonradiative decay. The luminescence transients of both GaN: Er samples were measured at 15 K, 300 K, and 520 K. It was observed that the decay curves were non-exponential which suggests the existence of multiple Er sites. The existence of multiple Er sites in GaN has been previously reported [11]. To describe the lifetime decay an average lifetime was used. For GaN: Er (SSMBE) it was observed that the low temperature (15 K) lifetime of 2.3 ms decreased to 1.9 ms at room temperature. At higher temperatures the lifetime continued to decrease and reached a value of 1.2 ms at 520 K. The decrease of the lifetime above 300K is most likely due to the onset of nonradiative decay. Compared to GaN: Er (SSMBE), the lifetime of GaN:Er (MOMBE) was significantly shorter and decreased slightly from 0.11 ms at 15 K to 0.10 ms at room temperature. At higher temperatures the lifetime was too short to be measured with our current setup. The room temperature luminescence efficiencies were estimated from the ratio of the low and room temperature lifetimes (τ_{300K}/τ_{15K}) to be ~0.8 for GaN: Er (SSMBE) and ~0.9 for GaN: Er (MOMBE),

respectively. The high PL efficiencies indicate that the Er^{3+} excitation efficiency and the concentration of Er^{3+} ions limit the device performance of current infrared LED's.

The visible PL spectra of the GaN: Er samples following optical excitation at 325 nm are shown in Figure 3. The GaN: Er (SSMBE) exhibited a weak bandedge PL at ~369 nm (3.36 eV) and two "green" lines located at 537 nm and 558 nm. The green luminescence was assigned to the intra 4f Er^{3+} transitions $^2H_{11/2} \rightarrow {}^4I_{15/2}$ and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ (Ref. 3). The GaN: Er (MOMBE) sample showed strong bandedge PL located at ~381 nm (3.25 eV), however, no indication of green Er^{3+} luminescence was found. As discussed before, for GaN: Er (MOMBE) the bandedge provides an efficient radiative combination channel, which reduces the excitation efficiency for both infrared and visible Er^{3+} transitions. Figure 3b) shows the decay transients of the green Er^{3+} PL at different temperatures. The lifetime was found to be non-exponential at all temperatures and decreased with increasing temperature. The average lifetimes for the 558 nm line at 15 K and room temperature were determined to be 10.8 µs and 5.5 µs, respectively.
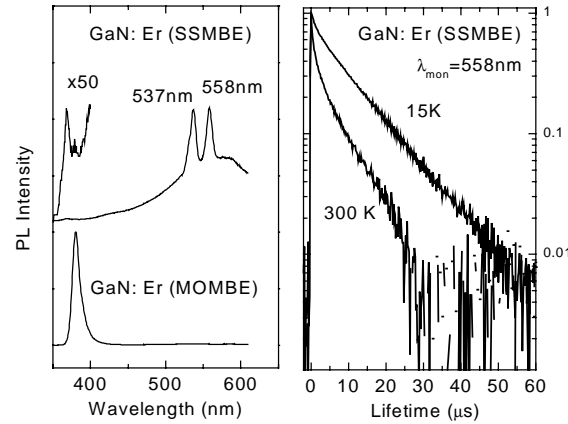

Figure 3: a) Visible PL spectra from GaN: Er (SSMBE) and GaN: Er MOMBE) at 300 K (λ_{ex}=325 nm). b) Decay transients of the visible PL at 558 nm from GaN: Er (SSMBE) at 15 K and 300 K (λ_{ex}=495 nm).

A more detailed study on the temperature dependence of the lifetime is depicted in Figure 4a). The thermalization of the $^4S_{3/2}$ and $^2H_{11/2}$ states leads to a common decay time τ (effective spontaneous emission probability), which can be described as:

$$\frac{1}{\tau} = \frac{\tau_S^{-1} + \tau_H^{-1} \cdot \frac{g_H}{g_S} \exp\left(\frac{-\Delta E}{kT}\right)}{1 + \frac{g_H}{g_S} \exp\left(\frac{-\Delta E}{kT}\right)} \tag{1}$$

where τ_H and τ_S are the intrinsic radiative decay times of the $^2H_{11/2} \rightarrow {}^4I_{15/2}$ and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transitions, respectively. g_H and g_S are the electronic degeneracies (2J+1) of the $^2H_{11/2}$ and $^4S_{3/2}$ states and ΔE is their energy difference (ΔE=87meV) .
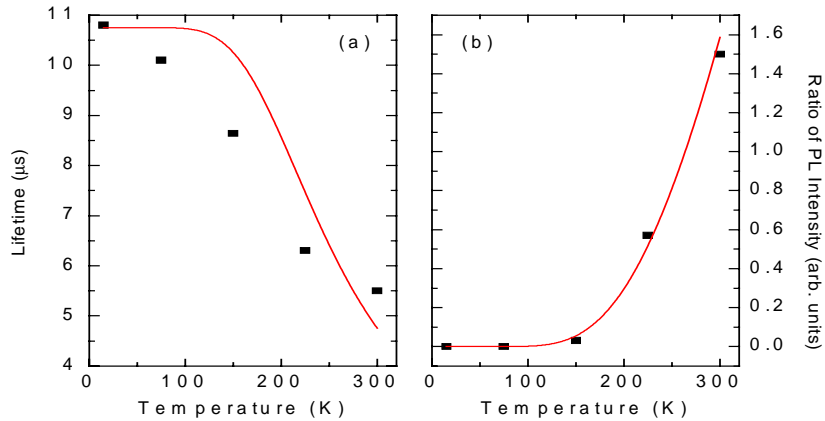
Figure 4: *a) Green PL lifetime of GaN: Er (SSMBE). The solid line describes the change in lifetime according to equation (1). b) Ratio of the PL intensities of the green lines as a function of temperature (see reference 3).*

At low temperatures the $^2H_{11/2}$ state is not thermally populated and the experimental lifetime can be approximated as the intrinsic decay time of the $^4S_{3/2} \rightarrow \ ^4I_{15/2}$ transition, i.e. τ_S=10.8 μs. It is assumed in this approximation that the low temperature decay time is purely radiative. The intrinsic lifetime of the $^2H_{11/2} \rightarrow \ ^4I_{15/2}$ is not experimentally accessible, unless a careful analysis of absorption data is carried out, which is rather difficult for thin film materials. It is possible, however, to obtain a rough estimation of the intrinsic decay time τ_H from the temperature dependence of the luminescence intensity of the green Er^{3+} lines at 537 nm and 558 nm. Steckl and Birkhahn [3] reported that the intensity of the $^4S_{3/2}$ line decreased with increasing temperature, whereas the $^2H_{11/2}$ line had a maximum of intensity at around 300 K. Taking the experimental data from reference 3, the ratio of the 537 nm and 558 nm lines was calculated and is plotted in Fig 4b). Considering the thermal coupling of the involved states, the intensity ratio of both lines was fitted to an expression

$$I_H = \frac{\tau_S \cdot g_H \cdot hw_H}{\tau_H \cdot g_S \cdot hw_S} \cdot \exp(-\Delta E / kT) \qquad (2)$$

with $h\omega_H$ =2.309eV, $h\omega_S$=2.222eV, τ_S=10.8 μs and ΔE=87 meV. τ_H was taken as a fitting parameter and the best fit to the data yielded τ_H=0.75 μs. The fitting result shows that the radiative rate of the $^2H_{11/2} \rightarrow \ ^4I_{15/2}$ transition is much larger than that of the $^4S_{3/2} \rightarrow \ ^4I_{15/2}$ transition, consistent with published data on Er doped insulators. Using this set of parameters the temperature dependence of the luminescence lifetime was calculated according to equation (1) and is shown in Fig. 4a). The modeling reveals that the decrease of the luminescence lifetime with temperature is mainly due to an increased radiative decay rate arising from the fast thermalization of the $^2H_{11/2} \rightarrow \ ^4I_{15/2}$ and $^4S_{3/2} \rightarrow \ ^4I_{15/2}$ transitions. This preliminary analysis of the lifetime implies that non-radiative decay processes are small and therefore the green luminescence efficiency is high. To obtain further support for

this conclusion it will be necessary to perform a more systematic study of the Er^{3+} visible PL lifetime for a series of samples with different Er concentrations.

SUMMARY

In summary, we performed a comparison of the infrared and visible PL properties of GaN: Er (MOMBE) and GaN: Er (SSMBE). We observed that both samples exhibited intense 1.54 μm PL under below-gap excitation. With above-gap excitation GaN: Er (MOMBE) showed a greatly reduced IR PL intensity, whereas the 1.54 μm PL from GaN:Er (SSMBE) remained strong. Based on temperature dependent PL intensity and lifetime studies, it was concluded that the 1.54 μm luminescence efficiency is high (~0.8-0.9). The factors limiting the performance of current IR LED's are the Er excitation efficiency and the Er concentration. No visible PL arising from intra-4f Er transitions was found from GaN: Er (MOMBE). On the contrary, the GaN: Er (SSMBE) sample revealed green lines at 537 nm and 558 nm with an average lifetime of 5.5 μs at room temperature. The temperature dependence of the green lifetime was explained by the strong thermalization of the $^2H_{11/2}$ and $^4S_{3/2}$ excited states. Non-radiative decay does not seem to affect the green luminescence efficiency.

ACKNOWLEDGMENTS

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