ELSEVIER

Journal of Alloys and Compounds 303–304 (2000) 331–335

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

Near infrared (1.54 μm) luminescence properties of erbium doped gallium nitride

U. Hömmerich[a,*], J.T. Seo[a], Myo Thaik[a], C.R. Abernathy[b], J.D. MacKenzie[b], J.M. Zavada[c]

[a]*Hampton University, Department of Physics, Research Center for Optical Physics, Hampton, VA 23668, USA*
[b]*University of Florida, Department of Materials Science and Engineering, Gainesville, FL 32611, USA*
[c]*U.S. Army European Research Office, London NW1 5TH, UK*

Abstract

The photoluminescence (PL) properties of in-situ Er doped GaN prepared by metalorganic molecular beam epitaxy (MOMBE) have been investigated. The GaN:Er films were grown on sapphire or silicon substrates. The oxygen and carbon background concentrations in the films were measured to be in the order of $\sim 10^{20}$ and $\sim 10^{21}$ cm^{-3}, respectively. Both types of GaN:Er samples showed intense 1.54 μm PL at room temperature under below-gap optical excitation. For above-gap excitation, a greatly reduced Er^{3+} luminescence intensity was observed. Pump intensity dependent PL studies revealed that the Er^{3+} excitation efficiency under above-gap excitation is roughly a factor of ~30 smaller compared to below-gap excitation. Based on the efficient Er^{3+} below-gap excitation process a hybrid GaN:Er/sapphire/InGaN LED was developed.

Keywords: Photoluminescence; Erbium doping; Gallium nitride

1. Introduction

Erbium (Er) doped semiconductors are currently being considered as potential near infrared light sources for optoelectronic applications [1–3]. It is well known that Er^{3+} ions exhibit atomic-like, temperature stable luminescence at 1.54 μm due to the intra-4f-shell Er^{3+} transition $^4I_{13/2} \rightarrow {}^4I_{15/2}$. Since the 1.54 μm emission from Er^{3+} overlaps the minimum absorption region of silica based optical fibers, Er doped semiconductors are suitable light sources for applications in optical communications. In contrast to existing 1.54 μm sources which are based on optically excited Er^{3+} ions in insulating materials or on the temperature sensitive bandedge emission from semiconductors, Er doped semiconductors offer the prospect of electrically pumped, compact, and temperature stable optoelectronic devices [3].

The photoluminescence (PL) properties of Er doped silicon and several III–V materials have been extensively studied for over a decade, however only weak Er^{3+} PL was observed at room temperature [1,2]. It was first reported by Favennec et al. that the Er^{3+} PL intensity depends strongly on both the bandgap energy of the semiconductor and the host temperature [4]. These researchers observed that the thermal quenching of Er^{3+} PL decreased for semiconductor hosts with larger bandgap. Therefore, current research activities focus on investigating Er^{3+} ions doped into wide-gap semiconductors [2,3].

The recent development of blue light emitting diodes and lasers based on GaN has stimulated an enormous amount of activity in the field of III-nitride semiconductor research [5,6]. The bandgap of III-nitrides ranges from 1.9 eV (InN) to 6.2 eV (AlN), which makes these materials also interesting candidates for Er^{3+} doping. Intense room temperature luminescence at 1.54 μm has been reported from Er doped GaN and AlN [7–11]. The first demonstration of electroluminescence from Er implanted GaN was reported by Torvik et al. in 1996 [12]. Subsequently, other researchers have reported infrared and visible electroluminescence from Er doped GaN [13]. These results underlined that III-nitrides are suitable Er^{3+} (and possibly other rare earth ions) host materials for electroluminescent devices operating at room temperature. However, the initial reports also indicated that much more spectroscopy and materials research is necessary to fully explore the incorporation and excitation mechanisms of Er^{3+} ions in III-nitrides. In this paper we present new spectroscopic

*Corresponding author. Fax: +1-757-727-5955.
E-mail address: hommeric@las.org (U. Hömmerich)

0925-8388/00/$ – see front matter
PII: S0925-8388(00)00602-2

results on the 1.54 μm luminescence properties of in-situ Er doped GaN samples grown on sapphire and silicon substrates and discuss their potential for optoelectronic applications. The development of a novel hybrid light emitting diode based on GaN:Er/sapphire/InGaN is also presented.

2. Experimental considerations

The Er doped GaN films were grown by metalorganic molecular beam epitaxy (MOMBE) in an INTEVAC Gas Source Gen II on In-mounted (100) silicon or (0001) sapphire substrates [14]. The GaN films were preceded by a low temperature AlN buffer ($T_g = 425$°C). A 0.2 μm undoped GaN spacer was deposited prior to the growth of GaN:Er. Triethylgallium (TEGa) and dimethylethylamine alane (DMEAA) provided the group III fluxes. A shuttered effusion oven with 4N Er was used for solid source doping. Reactive nitrogen species were provided by a SVT radio frequency plasma source. Due to the incorporation of carbon and oxygen from residual ether in TEGa, the C and O background observed were $\sim 10^{21}$ and $\sim 10^{20}$ cm^{-3}, respectively, as determined by SIMS measurements. PL measurements were carried out using either the visible (442 nm) or UV (325 nm) outputs of a HeCd laser. For pump power dependent studies the ultraviolet (333.6–363.8 nm) or visible lines (457–514 nm) of an argon laser were employed. Photoluminescence excitation (PLE) studies were performed using either an argon pumped dye laser or a Nd:YAG/Optical Parametric Oscillator system. Luminescence spectra and lifetime data were recorded using a 1-m monochromator equipped with a liquid nitrogen cooled Ge detector. The PL spectra were recorded using a standard lock-in technique. Luminescence lifetime data were taken using a digital oscilloscope. The samples were cooled on the cold finger of a two-stage closed-cycle helium refrigerator.

3. Experimental results and discussion

3.1. PL spectra under above- and below-gap Er^{3+} excitation

The room temperature infrared Er^{3+} PL spectra of GaN:Er/sapphire and GaN:Er/Si for above- and below-gap excitation are shown in Fig. 1. The samples were excited with either the 325 or 442 nm outputs of a HeCd laser. To avoid any saturation effects, the pump intensity was chosen to be less than 0.4 W/cm^2. The Er^{3+} PL properties under above-gap optical pumping are very important for future device development because the Er^{3+} excitation mechanism is similar to carrier injection and subsequent recombination occurring in a forward-biased light emitting diode. Both Er doped GaN samples showed characteristic near infrared emission centered at 1.54 μm, which can be assigned to the intra-4f Er^{3+} transition $^4I_{13/2} \rightarrow {}^4I_{15/2}$. Independent of excitation wavelength, the luminescence spectra were relatively broad (FWHM ~25–35 nm) and featureless, which suggests a homogenous distribution of Er^{3+} sites in the host matrix. Similar Er^{3+} PL spectra have been observed from Er doped glass [15]. It should be noted that the GaN:Er/silicon sample also showed 1.54 μm PL at room temperature, which is of great interest for applications in silicon based optoelectronics. A significant difference was observed in the Er^{3+} PL intensity for below- and above-gap excitation. For GaN:Er/sapphire the peak Er^{3+} PL intensity at 1.54 μm decreased by roughly a factor of ~55 when changing the pump wavelength from 442 to 325 nm. A change in Er^{3+} PL intensity by a factor of ~10 was observed for GaN:Er/Si under identical experimental conditions. A similar PL reduction was reported for Er implanted GaN [16]. Comparison of the Er^{3+} PL intensity under above- and below-gap excitation suggests that only a weak electroluminescence can be expected from a forward-biased GaN:Er LED. It is not yet clear why the Er^{3+} PL intensity was so greatly reduced under above-gap excitation. Preliminary visible luminescence studies (experimental data and analysis will be published elsewhere) indicate that the bandedge provides another radiative recombination channel, which reduces the overall Er^{3+} excitation efficiency. More work is currently in progress to elucidate the correlation between infrared Er^{3+} PL intensity and near bandedge luminescence from Er doped GaN.

3.2. Pump intensity dependent photoluminescence study

The Er^{3+} PL intensity of GaN:Er/Si was studied as a function of pump intensity in order to gain more insight into the Er^{3+} PL properties. Fig. 2a shows the Er^{3+} PL intensity monitored at 1.54 μm as a function of pump intensity at room temperature under above- and below-gap excitation. It can be seen that under below-gap excitation the Er^{3+} PL intensity shows a nonlinear behavior, suggesting the onset of Er^{3+} PL saturation. In contrast, under above-gap excitation a nearly linear increase of the Er^{3+} PL intensity was observed up to pump intensities as high as ~100 W/cm^2. The linear increase indicates that only a fraction of Er ions has been excited, even at the high pump intensities.

The observed pump intensity behavior can be modeled using a simple three level Er^{3+} energy scheme. The excitation of Er^{3+} into a pump level occurs with an absorption cross-section (σ_{abs}) and is followed by a fast relaxation into the metastable level. Only a certain fraction (η_p) of the pump level population quickly ends in the metastable level. The product of absorption cross-section (σ_{abs}) and pump efficiency (η_p) is used in the following as the overall Er^{3+} excitation efficiency (σ_{ex}). After balancing the rates of excitation and deexcitation processes under

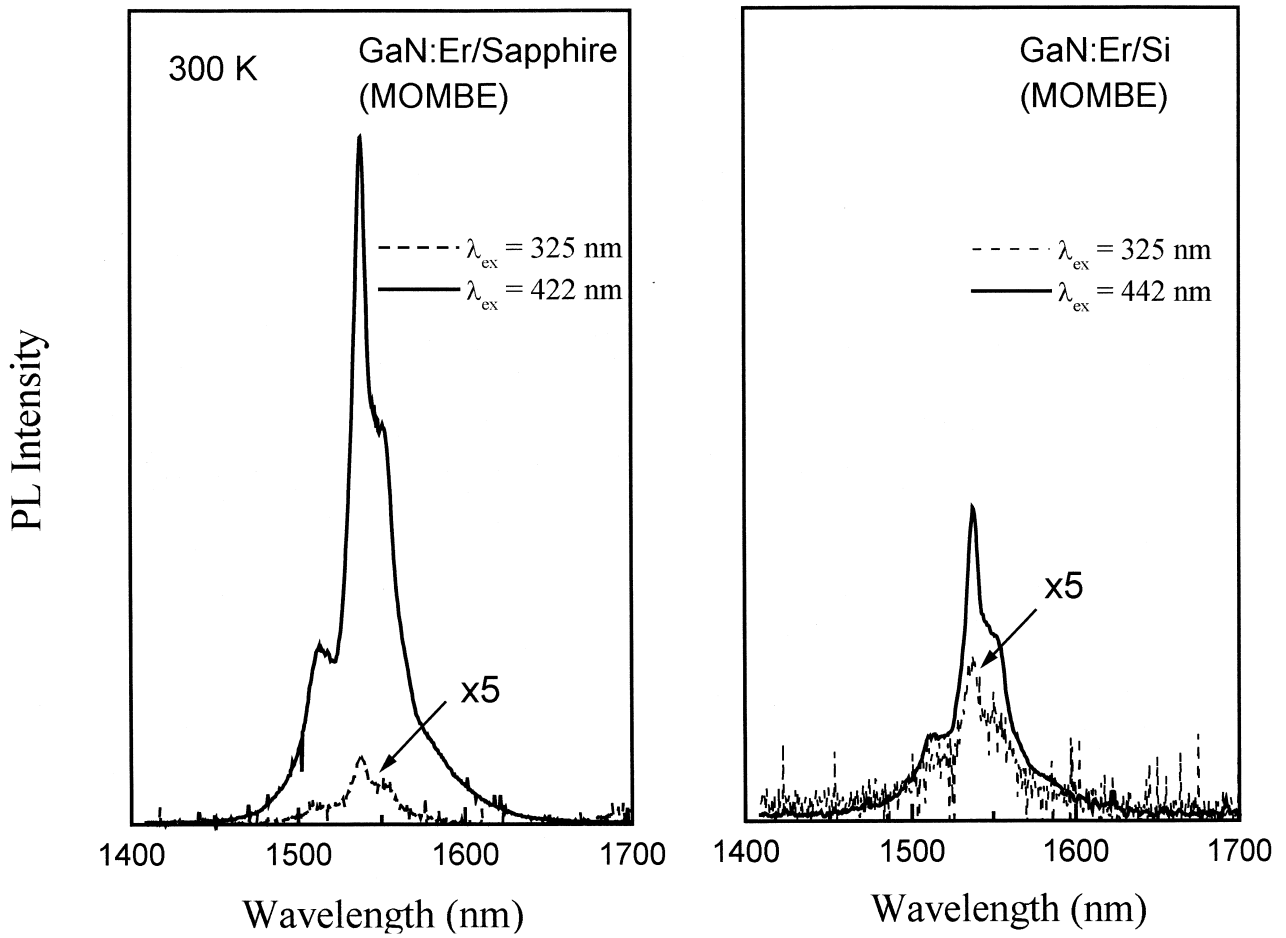

Fig. 1. Er^{3+} PL spectra from in-situ Er doped GaN on sapphire and silicon substrates under above- and below-gap excitation. All spectra were recorded at room temperature at a constant pump intensity of 0.4 W/cm^2.

steady state pumping, it can be derived that the pump power dependence of the Er^{3+} PL intensity is given as

$$I = \frac{N}{[1 + (hc/\tau \cdot \sigma_{ex} \cdot P \cdot \lambda_p)]} \cdot w_r \cdot C \quad (1)$$

where N is the concentration of optically active Er^{3+} ions, w_r is the radiative decay rate, C is the collection efficiency of the luminescence setup, τ is the luminescence lifetime, σ_{ex} is the excitation efficiency, λ_p is the excitation wavelength, and P is the pump intensity. In order to model the data shown in Fig. 2a, we assumed that the collection efficiency of the PL setup, the number of optically active Er^{3+} ions, and the radiative decay rate are independent of excitation wavelength. The luminescence transients for above- ($\lambda_{ex} = 355$ nm) and below-gap ($\lambda_{ex} = 532$ nm) excitation are depicted in Fig. 2b. For comparison, the system response is also shown in Fig. 2b. Within the experimental error of the measurement we observed that the lifetime does not depend on the excitation wavelength. The only remaining parameter used to fit the pump intensity data shown in Fig. 2b to Eq. (1) was the excitation efficiency. The obtained best fits revealed that the excitation efficiency under above-gap pumping is reduced by a factor of ~30 compared to below-gap excitation. As discussed before, preliminary visible luminescence studies have shown that the bandedge provides another radiative recombination channel, which reduces the overall Er^{3+} excitation efficiency under above-gap excitation. To what extent this reduction in excitation efficiency is due to a wavelength dependent absorption cross-section and/or pump efficiency is not yet known.

3.3. Development of a hybrid 1.54 μm GaN:Er/InGaN LED

The development of blue/green InGaN LEDs has made possible novel optoelectronic applications involving wavelength down conversion. These devices can be classified as ''hybrid LEDs'' because they are based on the optical excitation of light emitting centers using the electroluminescence from InGaN. A white light source was demonstrated by Nakamura using an InGaN LED to optically pump organic phosphor materials [17]. Another example of achieving white light emission was recently reported by Hide et al. [18]. These authors used an InGaN

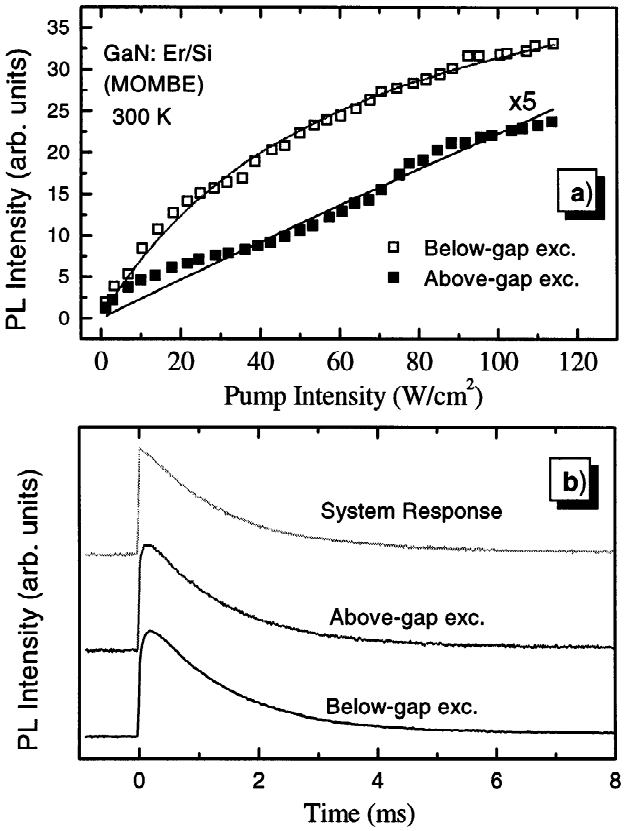

Fig. 2. (a) Er^{3+} PL intensity of GaN:Er/Si as a function of pump intensity at room temperature. A similar pump intensity behavior was observed for GaN:Er/sapphire. The solid line is the best fit to Eq. (1). (b) Decay transients of GaN:Er/Si under above- and below-gap excitation.

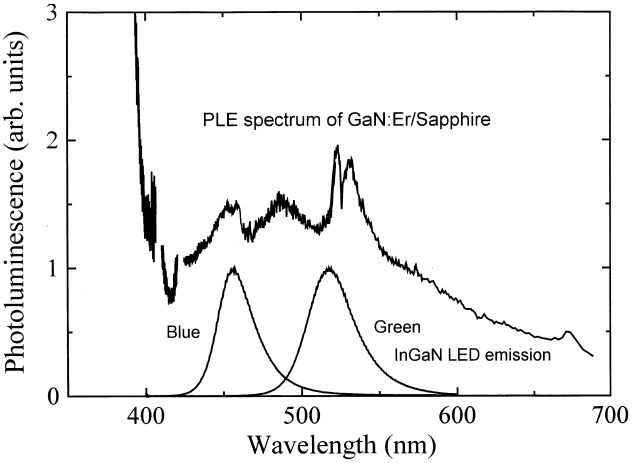

Fig. 3. Upper trace: photoluminescence excitation spectrum of GaN:Er/sapphire. Lower trace: blue and green emission spectra from a Nichia GaInN LED. The excellent overlap between the below-gap Er absorption band and the Nichia InGaN LED indicates the possibility of a ''hybrid'' LED based on Er doped GaN.

LED to optically excite conjugated polymers. Depending on the polymer film, the emission of this hybrid InGaN/conjugated polymer LED was tunable across the chromaticity diagram. As discussed before, our current GaN:Er films showed a significantly larger Er^{3+} excitation efficiency for below-gap excitation than for above-gap excitation. This observation has stimulated our interest in developing a hybrid LED based on Er doped GaN.

Using the technique of photoluminescence excitation (PLE) spectroscopy [19], we investigated the below-gap absorption bands of Er^{3+} in more detail and compared the result with the light emission from a commercial blue/green InGaN LED. It can be seen from Fig. 3 (upper trace) that Er^{3+} ions can be excited continuously at any wavelength between 400 and 600 nm. A similar below-gap Er absorption band has been observed for Er doped AlN [20]. The broad Er^{3+} absorption band nicely overlaps the output of an InGaN LED with emission bands peaking at 450 and 520 nm as shown in the lower trace of Fig. 3. These measurements indicate the possibility of a hybrid LED in which an Er doped GaN film is optically excited by a commercial blue/green InGaN LED to produce emission at 1.54 μm. Fig. 4 shows the design of a hybrid GaN:Er/sapphire/InGaN LED and its operation. In this experiment a blue/green Nichia InGaN LED was employed to excite the Er^{3+} ions in the GaN layer grown on a sapphire substrate. The epoxy bubble of the InGaN LED was removed and polished. The GaN:Er film was placed directly on top of this LED as shown in Fig. 4a. The DC forward voltage and current of the InGaN LED were 2.98 V and 20 mA, respectively. The luminescence at 1.54 μm under blue or green pumping was collected through the sapphire substrate and is displayed in Fig. 4b. Even though the resulting emission at 1.54 μm was weak, it demonstrates that the electroluminescence from InGaN can be used to excite Er^{3+} ions in GaN.

4. Conclusion

In conclusion, Er doped GaN films prepared by MOMBE emit intense 1.54 μm PL at room temperature when excited by below-gap radiation. Under above-gap excitation the Er^{3+} PL intensity decreased significantly. Pump intensity dependent Er^{3+} PL studies of GaN:Er/Si revealed that the Er^{3+} excitation efficiency for above-gap pumping is reduced by a factor of ~30 compared to below-gap pumping. The low above-gap excitation efficiency will be a limiting factor in current electroluminescence devices employing forward-biased carrier injection. Based on the efficient below-gap Er excitation a hybrid InGaN/GaN:Er LED operating at 1.54 μm has been demonstrated. Questions concerning the overall efficiency of this novel hybrid LED need to be further addressed in the future.

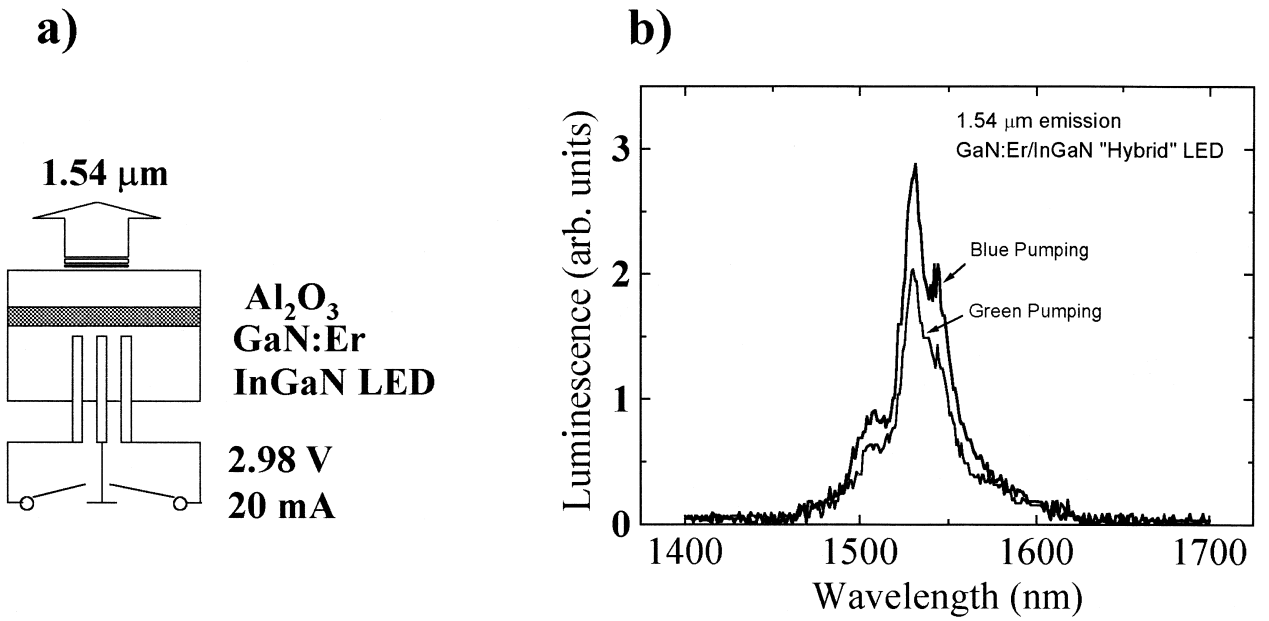

Fig. 4. (a) Design of a hybrid InGaN/GaN:Er LED. (b) Demonstration of 1.54 μm luminescence from the device shown in (a).

Acknowledgements

The authors from Hampton University acknowledge financial support from NASA through grant NCC-1-251 and the Army Research Office through grants DAAH04-96-1-0089 and DAAG55-98-1-0112. The work at the University of Florida was supported by ARO grant DAAH04-96-1-0089.

References

[1] G.S. Pomrenke, P.B. Klein, D.W. Langer (Eds.), Rare Earth Doped Semiconductors, Mater. Res. Soc. Symp. Proc. 301 (1993).
[2] S. Coffa, A. Polman, R.N. Schwartz (Eds.), Rare Earth Doped Semiconductors II, Mater. Res. Soc. Symp. Proc. 422 (1996).
[3] J.M. Zavada, D. Zhang, Solid State Electron. 38 (1995) 1285.
[4] P.N. Favennec, H. L'Haridon, D. Moutonnet, M. Salvi, Y. Le Guillou, Electron. Lett. 25 (1989) 718.
[5] S. Nakamura, T. Mukai, M. Senoh, J. Appl. Phys. 76 (1994) 8189.
[6] See MRS Bull. 22(2) (1997), Overview: GaN and Related Materials.
[7] R.G. Wilson, R.N. Schwartz, C.R. Abernathy, S.J. Pearton, N. Newman, M. Rubin, T. Fu, J.M. Zavada, Appl. Phys. Lett. 65 (1994) 992.
[8] C.H. Qiu, M.W. Leksono, J.I. Pankove, J.T. Torvik, R.J. Feuerstein, F. Namavar, Appl. Phys. Lett. 66 (1995) 562.
[9] J.D. MacKenzie, C.R. Abernathy, S.J. Pearton, U. Hömmerich, X. Wu, R.N. Schwartz, R.G. Wilson, J.M. Zavada, J. Cryst. Growth 175/176 (1997) 84.
[10] S. Kim, S.J. Rhee, D.A. Turnbull, X. Li, J.J. Coleman, S.G. Bishop, P.B. Klein, Appl. Phys. Lett. 71 (1997) 2662.
[11] R. Birkhahn, A.J. Steckl, Appl. Phys. Lett. 73 (1998) 2143.
[12] J.T. Torvik, J. Feuerstein, J.I. Pankove, C.H. Qiu, F. Namavar, Appl. Phys. Lett. 69 (1996) 2098.
[13] A.J. Steckl, M. Garter, R. Birkhahn, J. Scofield, Appl. Phys. Lett. 73 (1998) 2450.
[14] J.D. MacKenzie, C.R. Abernathy, S.J. Pearton, U. Hömmerich, J.T. Seo, R.G. Wilson, J.M. Zavada, Appl. Phys. Lett. 72 (1998) 2710.
[15] W.J. Miniscalco, J. Lightwave Techn. 9 (1991) 234.
[16] J.T. Torvik, R.J. Feuerstein, C.H. Qui, J.I. Pankove, F. Namavar, Mater. Res. Soc. Symp. Proc. 482 (1998) 579.
[17] S. Nakamura, G. Fasol, The Blue Laser Diode, Springer, 1997.
[18] F. Hide, P. Kozodoy, S.P. Den Baars, A. Heeger, Appl. Phys. Lett. 70 (1997) 2664.
[19] B. Henderson, G.F. Imbusch, Optical Spectroscopy of Inorganic Solids, Clarendon, Oxford, 1989.
[20] X. Wu, U. Hömmerich, J.D. MacKenzie, C.R. Abernathy, S.J. Pearton, R.N. Schwartz, R.G. Wilson, J.M. Zavada, Appl. Phys. Lett. 70 (1997) 2126.