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Characterization of InGaN structures grown by epitaxial lateral overgrowth over *a*-plane GaN template

SAULIUS MIASOJEDOVAS[1*], SAULIUS JURŠĖNAS[1], EDMUNDAS KUOKŠTIS[1], ARTŪRAS ŽUKAUSKAS[1], MIKHAIL E. GAEVSKI[2], JIAWEI LEE[2], MAXIM SHATALOV[2], ZHENG GONG[2], VINOD ADIVARAHAN[2], AJAY SATTU[2], IRINA MOKINA[2], JINWEI YANG[2], M. ASIF KHAN[2]

[1]Institute of Material Science and Applied Research, Vilnius University, Saultekio 9-III, LT-10222 Vilnius, Lithuania

[2]Department of Electrical Engineering, University of South Carolina, Columbia, SC 29208, USA

[*]Corresponding author: S. Miasojedovas, saulius.miasojedovas@ff.vu.lt

We report on the luminescence characterization of InGaN/GaN multiple quantum well (MQW) structures with average 15% In content in the well layers, grown on polar and non-polar sapphire substrates utilizing epitaxial lateral overgrowth (ELOG) technique. Significant modification of the emission properties of MQWs grown over non-polar ELOG structure in comparison with non-polar orientation was observed. It was attributed to the formation of In-rich quantum dot like structures in the vicinity of substrate related defects along stripes formed during ELOG procedure. The absence of the stimulated emission and the significant reduction of carrier lifetime, observed under strong excitation, indicate the high density of nonradiative centers in the In-rich quantum dot regions of non-polar ELOG MQWs.

Keywords: luminescence, InGaN/GaN, non-polar, multiple quantum wells (MQWs), epitaxial lateral overgrowth (ELOG), quantum dots (QDs).

1. Introduction

Group III-nitride semiconductors are the key materials for the blue and UV optoelectronic devices such as light emitting and laser diodes [1–3]. Recent progress in growth of nitride-based structures on non-polar substrates opens a new prospective in design of electronic and optoelectronic devices, without the restrictions imposed by strong internal electrostatic fields, which are always present in conventional *c*-plane structures [3–8]. However, these non-polar structures show high density of threading dislocations, due to insufficient control of the non-polar substrate quality, which leads to enhanced nonradiative recombination [3]. Epitaxial lateral overgrowth (ELOG) technique enabled significant reduction of the density of threading dislocations and an increase in carrier lifetime [3]. ELOG structures possess stripes with areas containing defects. Growth of the InGaN/GaN active structures on ELOG template, especially epilayers with sufficient content of In, can result in formation of In-rich regions in the surrounding of the template-related defects [9], what can remarkably alter optical properties of the device. Here we present luminescence characterization of InGaN/GaN multiple quantum wells (MQWs) with high In content grown over non-polar *a*-plane ELOG templates. High intensity photoexcitation condition applied, enabled carrier lifetime characterization under conditions close to semiconductor laser operation regime.

2. Experimental

The samples were prepared using low-temperature metal organic chemical vapor deposition (MOCVD) technique. The top layers of the samples with both *c*- and *a*-plane nitride quantum structures were fabricated using in both cases identical procedure, namely combining periodically changed precursors trimethylindium and trimethylgalium with constant flow of ammonia. The temperature was 675°C. However the templates with *a*- and *c*-plane orientation of GaN surface were prepared in different way. In the case of *c*-plane orientation, we used typical MOCVD regime for GaN growth over (0001)-surface (*c*-plane) sapphire wafer [10]. Under these circumstances InGaN/GaN MQW structure of up to 10 quantum wells (QW) was formed. In the case of *a*-plane GaN, we had to employ more complicated procedure in order to fabricate applicable smooth surface layer. First the *a*-plane GaN templates were grown over the *r*-plane sapphire with two-step low-pressure MOCVD using triethylgalium along with ammonia as precursors. The template GaN film thickness was close to 1 μm. Subsequently, an ELOG stripe mask was defined by 0.2 μm thick SiO_2 grown using plasma enhanced chemical vapor deposition. The mask pattern consisted of 12 μm width stripes and 6 μm width windows, which were opened using standard photolithography and wet etching with $HF:H_2O_2$. The SiO_2 mask stripes were oriented along the $[1\bar{1}00]$ direction. The template was then reloaded in the low-temperature MOCVD system, and additional growth was carried out at 1080°C for three hours and 1050°C for two hours, respectively. The lower temperatures enhanced the lateral growth rate, which resulted in a full coalescence of the GaN growth stripes. Nominal indium content in quantum wells was studied by means of energy dispersive X-ray spectroscopy and was about 15–18%. More detailed information on the growth conditions may be found elsewhere [11, 12].

The samples were excited by the fourth harmonic (photon energy $h\nu = 4.66$ eV) of the actively-passively mode-locked $YAG:Nd^{3+}$ (yttrium aluminum garnet) laser (pulse duration of $\tau = 20$ ps, repetition rate 10 Hz, maximum pump power per pulse 30 μJ). The size of excitation spot was approximately 1.5 mm. Luminescence was collected in backward geometry and dispersed by a 0.4 m grating monochromator. Toluene optical Kerr shutter was used for temporal resolution (20 ps) of the luminescence. The experiments were carried out at room temperature.

3. Results and discussion

The surface analysis of the samples with different crystallographic orientation by means of scanned electron and atomic force microscopy (AFM) revealed typically smooth surface for *c*-plane orientation whereas numerous dot-like heterogeneities with the dimension of 20–30 nm were obtained for *a*-plane samples. The AFM images of the a-plane surface before and after the final stage of deposition of InGaN-GaN are illustrated in Fig. 1. As can be seen these dots are definitely related with the top-layer fabrication procedure.

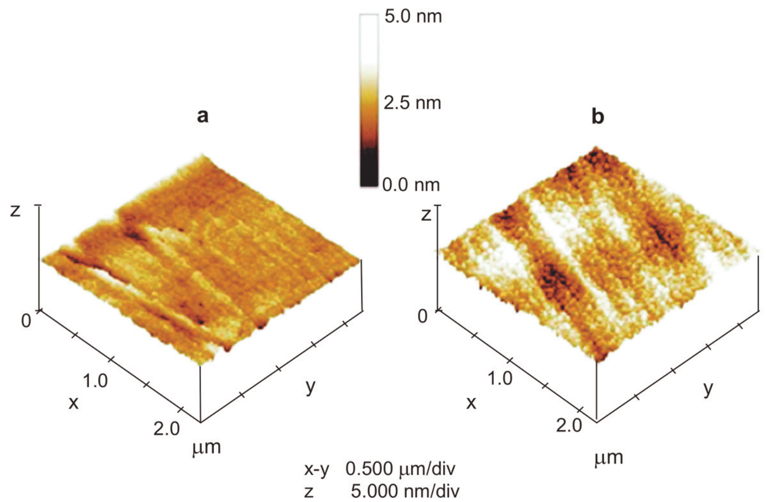

Fig. 1. AFM images of the *a*-plane sapphire before (**a**) and after (**b**) the final stage of deposition of InGaN/GaN.

Figure 2 depicts time-integrated luminescence spectra of InGaN/GaN MQWs grown on polar and non-polar ELOG templates, recorded at various excitation intensities. At low excitations, structure grown on polar *c*-plane showed conventional luminescence spectra with typical broad spontaneous luminescence band at 2.65 eV which originates from the QW regions. At high excitations ($I_p > 0.2$ mJ/cm^2), onset of stimulated emission at 2.91 eV on the high-energy wing is evidently seen.

Meanwhile structure grown on non-polar *r*-plane sapphire at low excitation intensities exhibited broad blueshifted luminescence line at 2.85 eV originated from the QWs. Blueshift of this band in non-polar structure can be the result of the reduced built-in electric field effect and/or decrease of indium concentration in the active layer. Besides, a new specific luminescence band appeared at about 3.23 eV, which can

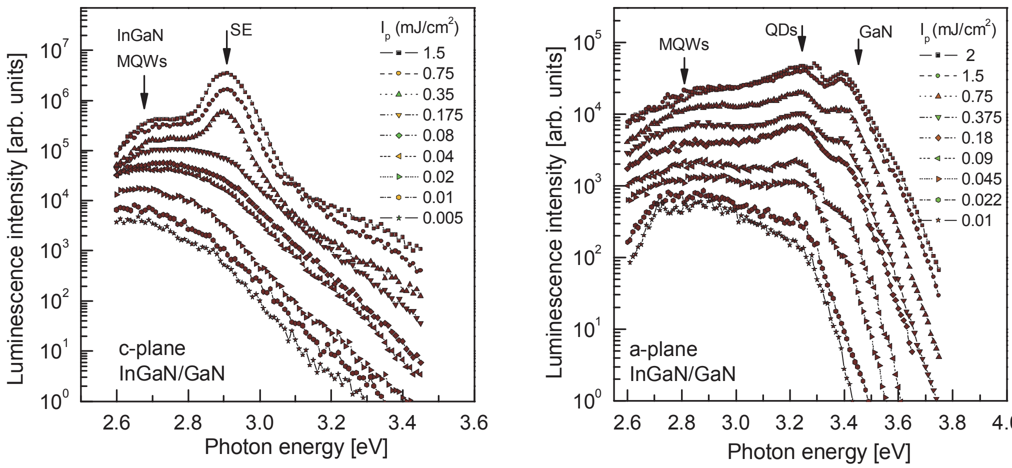

Fig. 2. Time-integrated luminescence spectra of InGaN/GaN MQWs grown on *c*-plane sapphire and on *r*-plane sapphire obtained at various excitation intensities.

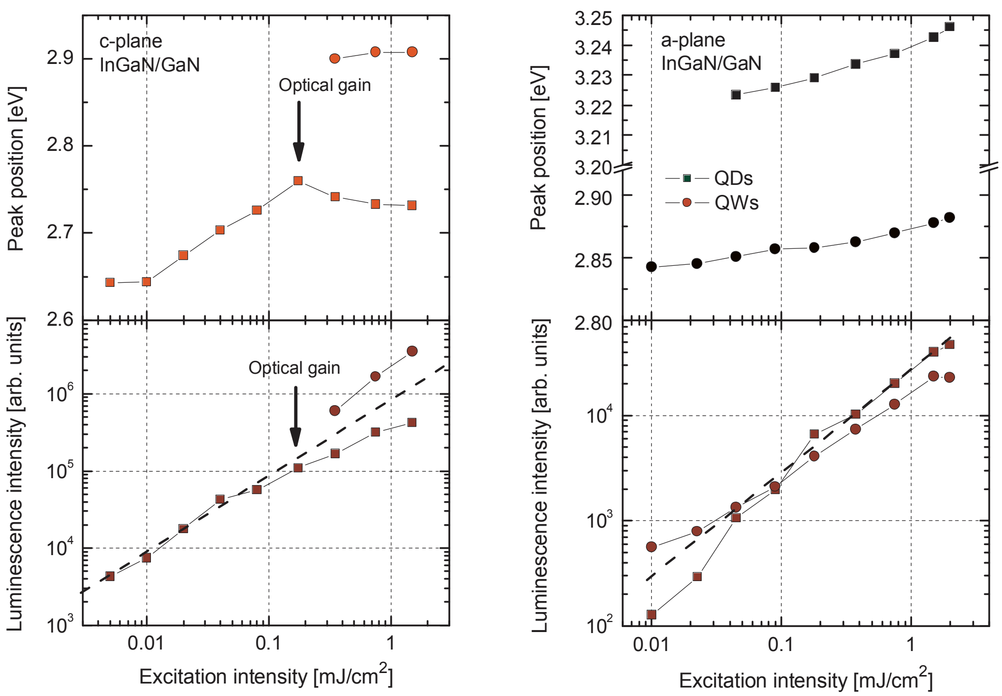

Fig. 3. Variation of spectra peak position and time-integrated luminescence intensity on excitation intensity of InGaN/GaN MQWs grown on *c*-plane sapphire (left hand side) and on *r*-plane sapphire (right hand side).

be related to luminescence from the In-rich quantum dot-like (QD) structures observed in the AFM images in Fig. 1. With the increase of excitation intensity, impact of QD luminescence increases. Spectra also contain less intensive luminescence band at 3.4 eV which is typical GaN band edge emission. Under the highest excitations stimulated emission at 3.3 eV from GaN layers appeared, but there was no stimulated emission from QDs, possibly due to high dislocation density in the structure.

Figure 3 shows variation of spectra peak position and integrated luminescence intensity on excitation intensity of polar (left hand side) and non-polar (right hand side) MQWs. With the increase of excitation intensity luminescence peak of polar structure of QWs blueshifts about 120 meV due to band filling and screening of built-in electric field. Under the highest excitation the built-in field can be regarded as completely screened by the carriers of high density. At about 0.2 mJ/cm^2 optical gain appears. Luminescence intensity of polar structure increases linearly with increase of excitation intensity. Appearance of optical gain is proved by superlinear luminescence intensity dependence on excitation intensity.

Luminescence band of non-polar MQW structure with increase of excitation intensity blueshifts about 40 meV, and QDs band blueshifts about 25 meV due to band filling effect. Meanwhile GaN luminescence band redshifts with increase of pump intensity due to band gap renormalization (not shown).

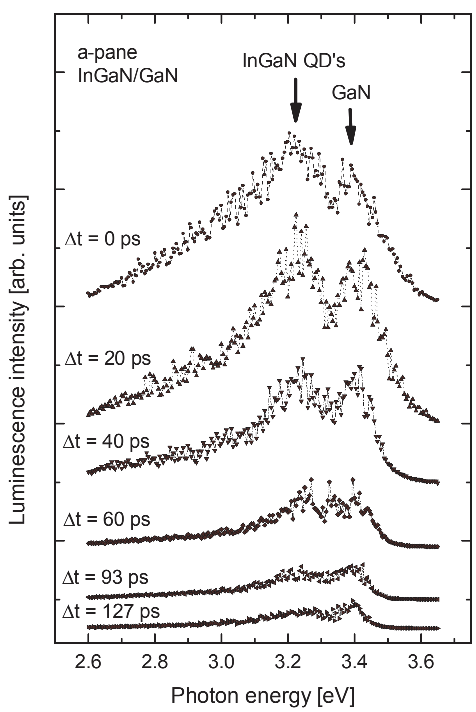

Fig. 4. Time-resolved luminescence spectra of InGaN QDs grown on *r*-plane sapphire. Spectra are arbitrary vertically shifted for clarity.

Figure 4 depicts luminescence spectra taken from non-polar structure at different delay times. Initially, after short pulse excitation, nonhomogeneously broadened band from QDs region and less intensive band from GaN layer side dominate the spectrum. With time, luminescence spectra narrows and no luminescence peak shift was observed. Spectra with temporal resolution of polar structure (not shown) show intensive stimulated luminescence during the first 100 ps and later only weak spontaneous luminescence can be resolved.

Figure 5 shows spectral dependencies of the late luminescence decay time for two structures. Polar structure shows typical increase of the luminescence decay time from 200 ps at the peak position to 700 ps for photons with lower energy. This indicates involvement in the recombination process of the deeper localized states. Decrease of the luminescence decay time in the energy region 2.95 eV is due to stimulated emission with fast depopulation of the states. Structure grown on non-polar ELOG substrate,

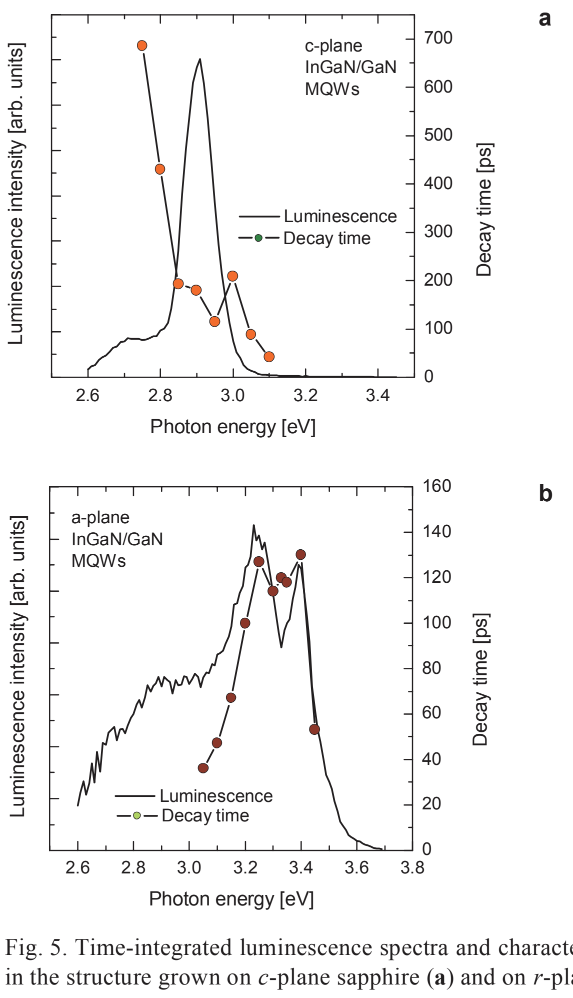

Fig. 5. Time-integrated luminescence spectra and characteristic decay time at different photon energies in the structure grown on *c*-plane sapphire (**a**) and on *r*-plane sapphire (**b**).

shows significantly shorter luminescence decay time $\tau = 130$ ps at QD and GaN regions. Decrease of the luminescence decay time for QD band can be attributed to the substrate-related defects.

4. Conclusions

In conclusion, InGaN/GaN MQWs grown on polar and non-polar ELOG substrates were characterized by luminescence spectroscopy under high excitation conditions, close to semiconductor laser operation regime.

Although polar structure suffers from built-in electric field, but at high excitation intensities built-in field is screened-up by free carriers and structure shows effective stimulated emission. Carrier lifetime in polar structures depends on delocalization level and it is twice longer in comparison with non-polar structure. Non-polar structures grown on ELOG template with high indium content posses self-formed indium-rich QDs, which give blueshifted luminescence. However QDs originate from areas containing defects with strong nonradiative recombination, which reduces carrier lifetime and suppress stimulated emission.

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