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Exciton localization in semipolar (1122) InGaN multiple quantum wells

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The exciton localization in semipolar (1122) InGa1−xN (0.13 ≤ x ≤ 0.35) multiple-quantum-well (MQW) structures has been studied by excitation power density and temperature dependent photoluminescence. A strong exciton localization was found in the samples with a linear dependence with In-content and emission energy, consistent with the Stokes-shift values. This strong localization was found to cause a blue-shift of the MQW exciton emission energy at temperature above 100 K, which was found to linearly increase with increasing In-content. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4960348]

I. INTRODUCTION

The major achievements in InGaN quantum-well (QW) active region based light-emitting diodes (LEDs) have mainly been made on the (0001) polar surface.1 However, this surface has a disadvantage, namely, the quantum-confined Stark effect (QCSE) caused by polarization induced electrostatic fields.2,3 This QCSE causes a strong spatial separation of electron and hole wave-functions resulting in low radiative recombination rates, which consequently reduce the internal quantum efficiency (IQE). The strength of QCSE decreases with increasing In-content due to the reduction in the effective field resulting in an enhanced luminescence efficiency as observed for polar QWs.2,15 The ELOC will cause a change in the carrier dynamics with increasing temperature resulting in an anomalous emission behaviour, namely, the S-shaped (“red-blue-red” shift) temperature dependence of QW emission energy that is usually attributed to hopping of excitons through the localized states.16,17 It should be noted that the ELOC intrinsically exists in InGaN alloys, irrespective of growth orientation employed.2,3,11,13–15,18–21 Zhang et al.14 have reported that (1122) QWs exhibit larger localization depths than polar QWs (estimated from TR-PL measurements at 7 K). Recently, by temperature-dependent photoluminescence (TD-PL) measurements, Zhang et al.14 demonstrated that the ELOC degree has been found in (1122) In0.2Ga0.8N QWs that causes a blue-shift of the QW exciton emission with rising temperature from ~200 K to 340 K, irrespective of excitation source used.21 However, so far there has been no systematic study on the ELOC in (1122) InGa1−xN QWs with different In-contents using TD-PL. The most likely reason is due to the high-TDD and high basal-plane stacking fault (BSF) density GaN templates grown on low-cost sapphire substrates that hinder the QW growth. Within the ALIGHT project,22 we have recently developed low-TDD low-BSF-density (1122) GaN templates grown on patterned (1012) r-plane sapphire substrates (up to 100 mm diameter).23

In this paper, by using TD-PL measurements with resonant excitation source, we systematically study on the ELOC in (1122) InGa1−xN (0.13 ≤ x ≤ 0.35) multiple-quantum-well (MQW) structures grown on high quality (1122) GaN templates on patterned r-plane sapphire substrates. Such an approach allows an exploration of general trends of the ELOC in (1122) InGa1−xN MQW.

II. EXPERIMENTAL DETAILS

The MQW samples investigated in this study were grown in a 3 × 2-in. Aixtron metal-organic vapour phase epitaxy (MOVPE) reactor on 6 μm-thick (1122) GaN templates.
that were prepared on patterned r-plane sapphire substrates (TDD \(\sim 2 \times 10^8 \text{ cm}^{-2}\), BSF density \(\sim 1 \times 10^{15} \text{ cm}^{-3}\)). The MOVPE growth of the templates is reported elsewhere.\(^{23}\) The MQW samples consisted of a 1.5 \(\mu\text{m}\)-thick Si-doped GaN layer, an active MQW region consisting of five periods of InGaN/GaN with nominal 2-nm-thick wells and 10-nm-thick undoped barriers. To investigate the effect of surface morphology of the templates on the ELOC in the MQW samples, few samples were also grown on chemically mechanically polished (CMP)-templates. Details of the MQW growth procedure and CMP process are described elsewhere.\(^{24}\) The surface morphology of the samples was investigated by Veeco MultiMode atomic force microscope (AFM) in tapping mode. The In-content of the samples was estimated using simulations based on x-ray diffraction measurements.\(^{21,24}\) To avoid the carrier diffusion problem from GaN to InGaN, TD-PL measurements of the samples have been performed using a continuous-wave violet laser diode \((E_{\text{ex}} = 3.06 \text{ eV})\) with excitation power densities \((P_{\text{ex}})\) of 2–15 W/cm\(^2\). The samples were mounted in a closed-cycle cryostat equipped with a Sumitomo (SHI) Cryogenics air-cooled compressor. The PL emission spectra were measured using a Horiba iHR320 spectrometer equipped with a monochromator and a thermoelectrically cooled Synapse CCD detector. The temperature measurements were varied from 10 K to 480 K. Additionally, PL and PL-excitation (PLE) measurements of the samples were also performed at 10 K to investigate the Stokes-shift using a monochromator coupled continuous-Xenon-lamp \((P_{\text{ex}} \sim 10^{-5} \text{ W/cm}^2)\). In this case, a photomultiplier tube was used to detect the PL signal with a lock-in amplifier system.

III. RESULTS AND DISCUSSION

The typical AFM images of the MQW samples grown on as-grown and CMP-templates are shown in Fig. 1. All samples show undulated surface morphology along [1100]. This typical morphology has been commonly observed on nonpolar and semipolar (Al,In,Ga)N surfaces.\(^{6,21,24–28}\) This can be well explained the anisotropic diffusion of group-III atoms on these surfaces.\(^{26,27,29}\) The sample grown on a CMP template (Fig. 1(b)) shows smoother morphology with a root-mean square (rms) roughness of 3 nm for a scan size of 20 \(\times\) 20 \(\mu\text{m}^2\), compared to an rms value of 25 nm estimated for the sample grown on an as-grown template (Fig. 1(a)).

For example, Fig. 2(a) shows TD-PL spectra of a (1122) InGaN MQW sample \((x = 16\%)\) grown on an as-grown template. All the spectra were fitted using a Gaussian function. As shown in Fig. 2(b), the MQW emission energy shows a non-monotonous peak shift behaviour (i.e., an S-shaped temperature dependence), irrespective of \(P_{\text{ex}}\) used. The MQW emission energy starts to exhibit a blue-shift at above a switching temperature \((T_{R\rightarrow BL})\) of \(\sim 160\) K, then exhibits a red-shift at \(T_{BL\rightarrow R} \geq 280\) K. This can be explained via the hopping processes of excitons through the localized states.\(^{2,3,15–18,21}\) When temperature increases from 10 K to \(T_{R\rightarrow BL}\), weakly localized carriers are thermally activated and they hop towards other strongly localized states resulting in the initial red-shift of emission energy. With increasing temperature from \(T_{R\rightarrow BL}\) to \(T_{BL\rightarrow R}\), localized carriers at lower energy levels will be strongly thermally activated to higher levels leading to the blue-shift of emission energy. Further increasing temperature above \(T_{BL\rightarrow R}\), the emission energy decreases following the Varshni law as this decrease is based on thermally induced changed in lattice constants and

\[ E_{\text{ex}} = 3.06 \text{ eV} \]
\[ P_{\text{ex}} = 15 \text{ W/cm}^2 \]

\[ x = 0.16 \]

\[ E_{\text{ex}} = 3.06 \text{ eV} \]
\[ P_{\text{ex}} = 15 \text{ W/cm}^2 \]

\[ x = 0.16 \]
\[ P_{\text{ex}} (\text{W/cm}^2) \]

\[ T_{R\rightarrow BL} \]
\[ T_{BL\rightarrow R} \]

\[ T_{R\rightarrow BL} \]
\[ T_{BL\rightarrow R} \]
binding strength.\(^\text{30}\) It should be noted that the sample \((x = 15\%)\) grown on a CMP-template also shows the same behaviour, despite the much smoother surface morphology (Fig. 1).

A similar behaviour has been found for other samples, irrespective of template used. An example can be seen in Ref. 21 for \((11\overline{2}2)\) In\(_{0.2}\)Ga\(_{0.8}\)N MQW. The switching temperatures (i.e., \(T_{R \to BL}\) and \(T_{BL \to R}\)) as a function of In-content are shown in Fig. 3. Both temperatures monotonically increase with increasing In-content indicating that the ELOC will dominate at higher temperatures.

The temperature-induced blue-shift of the MQW emission energy can be described by the Gaussian type band tail mode\(^\text{15}\)

\[
E_g(T) = E_g(0) - \frac{\alpha \cdot T^2}{\theta + T} - \frac{\sigma_E^2}{k_B \cdot T},
\]

where \(E_g(0)\) is the bandgap energy of InGaN at 0 K, \(k_B\) is the Boltzmann constant, \(\alpha\) is the Varshni fitting parameter, \(\theta\) is the Debye temperature (\(\sim 800\) K for GaN), \(\sigma_E\) is the dispersion of the band tail, which is correlated to the ELOC degree, i.e., the larger value of \(\sigma_E\) means the stronger localization effect. Examples of fitted curves are shown in Fig. 2(b).

Figure 4 shows the fitted value of \(\sigma_E\) as a function of In-content and emission energy at 10 K. The \(\sigma_E\) value linearly increases from \(-40\) meV to \(-90\) meV with increasing In-content from 13% to 35%, irrespective of template used. Additionally, the \(\sigma_E\) values also show a linear dependence on the MQW emission energy (the inset of Fig. 4) and tend to a zero value around the GaN bandgap of 3.5 eV. This is consistent with an increased full-width at half maximum (FWHM) value of the 10 K PL spectrum with increasing In-content. For the 10 K FWHM values \((P_{ex} = 15\) W/cm\(^2\)) were estimated to be 60 meV, 125 meV, and 180 meV for the samples with the In-content of 13% \((E_{\text{emission}} = 2.982\) eV), 30% \((E_{\text{emission}} = 2.480\) eV), and 35% \((E_{\text{emission}} = 2.285\) eV), respectively. The \(\sigma_E\) values are comparable irrespective of different \(P_{ex}\) used. This is consistent with the data shown in Fig. 2(b) and a previous report for \((11\overline{2}2)\) In\(_{0.2}\)Ga\(_{0.8}\)N MQW.\(^\text{21}\) Since the measurements were performed under the weak \(P_{ex}\), the QCSE can be neglected. For the \((11\overline{2}2)\) samples studied here, the unchanged peak emission energies and \(\sigma_E\) values indicate that a strong ELOC exists in the samples. A similar finding has been previously reported for \(m\)-plane In\(_{1-x}\)Ga\(_x\)N MQW samples \((5\% < x < 30\%)\)\(^\text{18}\) though these samples grown on high-TDD high-BSF-density templates showed much less pronounced \(S\)-shaped behaviour compared to the samples studied here. The large \(\sigma_E\) value of the \((11\overline{2}2)\) samples studied here is believed to derive from the anisotropic growth on the \((11\overline{2}2)\) surface\(^\text{26,27,29}\) and the onset of relaxation in \((11\overline{2}2)\) nitrides.\(^\text{28,31}\) The strong ELOC in \(m\)-plane MQW\(^\text{18}\) and the \((11\overline{2}2)\) MQW samples studied here can be another reason that should be taken into account with the reduced polarization fields\(^\text{4,5}\) to explain why non-/semi-polar MQW structures have a much shorter radiative lifetime compared to polar MQW.\(^\text{4,12-14,20}\)

To investigate the Stokes-shift of the samples, PL spectra together with PLE absorption edge were measured at 10 K using the Xe-lamp excitation source \((P_{ex} \sim 10^{-5}\) W/cm\(^2\)). The PLE measurements were performed with a detection energy fixed at each MQW peak emission energy. Figure 5(a) shows examples of 10 K PL spectra of two samples measured with \(E_{\text{ex}} = 3.815\) eV. The GaN near-band-edge (NBE) luminescence at about 3.475 eV and GaN donor-acceptor-pair (DAP) at about 3.3 eV can be clearly seen, while the MQW NBE can be observed at about 2.6 eV and 2.7 eV. It should be noted that the FWHM value of the 10 K PL spectrum under the Xe-lamp excitation is comparable with the value obtained under the laser excitation, similar to a previous finding for \((11\overline{2}2)\) In\(_{0.2}\)Ga\(_{0.8}\)N MQW.\(^\text{21}\)

The PLE edge of the \((11\overline{2}2)\) InGaN/GaN MQW samples was estimated using a sigmoidal fit\(^\text{12,33}\)

\[
\alpha(E) = \frac{1}{1 + \exp\left(\frac{E_B - E}{\Delta E}\right)},
\]

\[\text{FIG. 4. The ELOC degree (}\sigma_E\text{) of the (11\overline{2}2) InGaN MQW samples grown on as-grown (open symbols) and CMP-templates (dot-centre symbols) as a function of In-content. The inset shows the ELOC degree as a function of the MQW emission energy measured at 10 K.}\]
with previous results estimated for (1122) GaN MQW samples and GaN are shown in Fig. 5(b) together with the trend (solid-lines) of (1122) InGaN MQW samples (this work) and (1122) GaN templates prepared on patterned r-plane sapphire substrates. A strong exciton localization (ELOC) was found in the samples which shows a linear dependence with In-content and emission energy, consistent with the Stokes-shift values. This strong ELOC was found to cause a blue-shift of the MQW exciton emission at temperature above 100 K. This temperature was found to increase with increasing In-content. Though the high quality templates were used, no improvement in optical efficiency was observed. This is attributed to a high concentration of unintentional impurities and point defects in the samples. It is therefore expected that the efficiency of (1122) InGaN MQW can be significantly improved by lowering these concentrations.

IV. CONCLUSIONS

In summary, TD-PL measurements were carried out on (1122) In$_{x}$Ga$_{1-x}$N MQW samples (0.13 $\leq x \leq 0.35$) grown on high quality (1122) GaN templates prepared on patterned r-plane sapphire substrates. A strong exciton localization (ELOC) was found in the samples which shows a linear dependence with In-content and emission energy, consistent with the Stokes-shift values. This strong ELOC was found to cause a blue-shift of the MQW exciton emission at temperature above 100 K. This temperature was found to increase with increasing In-content. Though the high quality templates were used, no improvement in optical efficiency was observed. This is attributed to a high concentration of unintentional impurities and point defects in the samples. It is therefore expected that the efficiency of (1122) InGaN MQW can be significantly improved by lowering these concentrations.

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See http://www.alight-project.eu for the ALIGHT project description.