Optical and Structural Properties of $\text{In}_{0.08}\text{GaN}/\text{In}_{0.02}\text{GaN}$ Multiple Quantum Wells Grown at Different Temperatures and with Different Indium Supplies

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The optical and structural properties of $\text{In}_{0.08}\text{Ga}_{0.92}\text{N}/\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ multiple quantum wells (MQWs) grown at different temperatures and with different supplies of indium were analyzed by atomic force microscopy and spectrally resolved cathodoluminescence (CL). By comparing the contrasts of monochromatic CL images with high-resolution secondary-electron images of the sample surface, it is shown that almost all contrasts of the CL images can be explained by lateral inhomogeneities of both the thickness and the InN mole fraction of the InGaN layers. Dark contrasts in the CL images solely related to dislocations were not observed, indicating very weak nonradiative recombination correlated with threading dislocations in the InGaN quantum wells. The lateral inhomogeneities of layer thickness and indium incorporation depend strongly on the growth conditions.

Key words: InGaN, quantum wells, optical properties, cathodoluminescence, defects, dislocations

INTRODUCTION

InGaN/(AlIn)GaN multiple quantum wells (MQWs) are widely used as active regions for semiconductor lasers in the violet and blue spectral range.^{1,2} Such lasers are mainly applied as light sources for high-density optical data storage but also find many other applications, for example, in spectroscopy and in the medical field. Nevertheless, for applications such as laser projection displays,³ which require high output powers, blue and green semiconductor lasers are still not available. The emission of InGaN/(AlIn)GaN-based lasers can be expanded to the green wavelength range by increasing the indium content in the quantum wells. However, the incorporation of high levels of indium into the quantum wells (QWs) while

maintaining good structural quality poses a great challenge for the crystal growth.⁴

One way to achieve higher indium concentrations in InGaN MQWs is reduction of the growth temperature (T_G) during metalorganic vapor-phase epitaxy (MOVPE).^{4,5} With higher indium content (x_{solid}) in the QWs, however, the interface strain in the layer stack increases, and therefore a higher defect density is expected, limiting the efficiency and reliability of such lasers.⁴ Therefore, the role of dislocations which can act as nonradiative recombination centers is of major interest in this material system. In the past, several papers have been published presenting different conclusions regarding the role of dislocations in recombination in InGaN.^{4–10} Mukai et al.⁶ found that the emission efficiency of InGaN light-emitting diodes (LEDs) does not differ when they are grown on epitaxial lateral overgrowth (ELOG) GaN or on sapphire, whereas the dislocation density differs significantly. This is supported by the theoretical works of

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Elsner et al.⁷ as well as Arslan and Browning,⁸ who found that none of the different types of dislocations in GaN introduce states in the bandgap. Therefore, dislocations can exhibit nonradiative recombination activity only due to point defects confined in the strain field around a dislocation.⁹ The point defect concentration depends strongly on the growth conditions and can differ greatly, especially depending on the amount of indium incorporated into the quantum well. On the other hand, Sugahara et al.¹⁰ found a strong correspondence of dark spots in monochromatic cathodoluminescence (CL) images from InGaN/GaN MQWs obtained at 300 K for the CL wavelength of both the GaN buffer layer and the InGaN QWs. They concluded that these dark spots correspond to threading dislocations in both materials, indicating their activity as nonradiative recombination centers. Also, Nagahama et al.⁴ showed that the threshold current density of InGaN laser diodes emitting at 450 nm grown on ELOG GaN substrates with low dislocation density is much lower than that of the same layer stack grown on substrates with higher dislocation density.

In this work, we studied the influence of different growth temperatures on the optical properties of $\text{In}_{0.08}\text{Ga}_{0.92}\text{N}/\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ MQWs. In a series of samples the thickness as well as the indium content (x_{solid}) of both wells and barriers were kept constant while varying T_{G} . To this end, the indium supply in the gas phase (x_{vapor}) had to be changed for each T_{G} . The surface structure of the samples was investigated by atomic force microscopy (AFM) and high-resolution scanning electron microscopy in secondary-electron (SE) mode. Structural properties such as layer thickness (t_{well}) and x_{solid} of the QWs were obtained by high-resolution x-ray diffraction (HRXRD). The optical properties were studied using low-temperature (6 K) and room-temperature (RT) cathodoluminescence (CL).

EXPERIMENTAL PROCEDURES

The samples were grown by metalorganic vapor-phase epitaxy (MOCVD) in an AIX2600 HT reactor using standard precursors. All samples were grown on (0001) sapphire substrates with a 1.8- μm -thick GaN buffer layer on top. The growth temperatures for the $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{In}_y\text{Ga}_{1-y}\text{N}$ active region were varied from 760°C to 840°C in order to determine which growth conditions result in the best crystalline quality of the QW layers. x_{vapor} was varied from 0.16 to 0.53 depending on T_{G} to ensure the same x_{solid} for all samples. The active region consisted of fivefold $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{In}_y\text{Ga}_{1-y}\text{N}$ MQWs with an indium content of $x_{\text{solid}} = 0.08$ in the QWs and QW thickness (t_{well}) of about 3 nm. The barrier layers were grown with an indium content of $y = 0.02$ and thickness of 7 nm. The growth was stopped at the topmost barrier layer. More detailed information about the growth conditions can be found elsewhere.¹¹

AFM surface inspection was done with a Digital Nanoscope 3 system in tapping mode. Average layer thicknesses and indium concentrations of both QW and barrier layers were determined by HRXRD from comparison of $\Omega/2\Theta$ scans collected using a PANalytical X'Pert system with simulations using Epitaxy software. CL investigations were carried out in a Zeiss Ultra55 scanning electron microscope equipped with a Gatan Mono-CL3 system. CL spectra and monochromatic CL images were acquired simultaneously with inspection of the surface by secondary electrons (SE). The accelerating voltage was chosen to be 5 kV, and the electron probe current was 200 pA. In this case the spectral resolution is 0.5 nm, and the CL excitation depth amounts to 120 nm. Thus, electron-hole pairs were created within all QWs as well as in the uppermost part of the underlying GaN buffer layer.

RESULTS AND DISCUSSION

First, surface inspection by AFM and SE was done, revealing the existence of growth spirals for all samples examined in this study (Fig. 1).¹² The spiral diameters were observed to be about 400 nm to 800 nm. The density of growth spirals was about $3 \times 10^7 \text{ cm}^{-2}$ and did not differ between samples grown at different temperatures. By AFM, local variations of the spiral domain height were observed, being largest (about 8 nm) for the lowest growth temperature of 760°C. At higher T_{G} the samples were smoother, resulting in height differences of about 4 nm at 840°C. Additionally, white spots were observed in both AFM and SE images for the 840°C sample. These spots probably represent indium droplets on the sample surface¹³ and are not considered in the following. Furthermore, all SE images showed a large number ($1 \times 10^9 \text{ cm}^{-2}$) of pits. Some of these pits were located in the center of a growth spiral, but most of them were randomly distributed. The pit density was equal to the overall threading dislocation density determined from the full-width at half-maximum (FWHM) of the Ω -scans at the (0002) and (30 $\bar{3}$ 2) reflections of the GaN buffer layer.¹⁴

Therefore, we conclude that these pits are the escape points of the threading dislocations. Also, the pit density does not change with T_{G} . This can be understood by taking into account that the dislocation density in the MQW samples is determined by two factors: the dislocation density in the GaN buffer layer and the strain due to the different lattice constants of InGaN and GaN. Since the average thickness and indium concentration of the QWs as well as the buffer layer are the same for all samples, also the strain energy is similar and thus the number of dislocations releasing the strain should be equal.

Figure 2 shows CL spectra of the samples, which were acquired while exciting a region of $30 \mu\text{m} \times 40 \mu\text{m}$ (over several tens of growth spirals) of the

Fig. 1. SE (a, c) and AFM (b, d) images of the surface of samples grown at 760°C and 840°C, respectively.

Fig. 2. CL sum spectra over an area of $30\text{ }\mu\text{m} \times 40\text{ }\mu\text{m}$ for samples grown at 760°C, 780°C, 820°C, and 840°C.

respective sample. For all samples, the peak wavelength was found to be about 405 nm. In the spectra, besides the main peak, additional peaks with a distance of about 95 meV are seen on the low-energy side, which we attribute to phonon replica of the main emission. The FWHM of the main peak in the spectra compared with the average spiral height found from AFM measurements is plotted against T_G in Fig. 3. The smallest FWHM is observed at 780°C. Despite the reduction of the spiral height with increasing T_G and thus the reduction of layer

Fig. 3. FWHM of CL sum spectra and average height differences in AFM images versus growth temperature.

thickness fluctuations, the FWHM increases again for temperatures $> 780^\circ\text{C}$.

Monochromatic 6-K CL images taken at different wavelengths are shown in Fig. 4 for the samples grown at the lowest (760°C) and highest temperature (840°C). The images were taken at the peak wavelength and at the wavelengths observed at half of the maximum intensity on the high- and low-energy slopes of the spectrum. In the images taken at the peak wavelength (Fig. 4a, b), dark contrast is visible inside the spirals (circles in Fig. 4a, b), which could be attributed to non-radiative recombination connected with the screw

Fig. 4. Monochromatic CL images (6 K) taken at the peak wavelength (a, b), the high-energy slope (c, d), and the low-energy slope (e, f) for the samples grown at 760°C and 840°C, respectively.

dislocation inside the spirals. However, in the images taken at the low-energy slope of the peak (Fig. 4e, f), the corresponding region appears bright. Note that at least one dislocation is found in the center of the spirals, as seen in the SE images. However, no dark spot corresponding to nonradiative recombination at those dislocations is found in the long-wavelength images. On the other hand, if the spectrum consisted of a single emission line only, the images taken at the peak wavelength as well as at the wavelengths on the high- and low-energy slopes of the spectrum should show the same intensity distribution. Since we observe a strong anticorrelation of intensities at different wavelengths we conclude that the spectrum taken over several tens of growth spiral domains is a

superposition of many single spectra from different lateral positions within the excited area, indicating a wavelength variation across the spiral domains. This was proven by taking CL spectra along a line over several growth spirals (Fig. 5). As an example, for the sample grown at 780°C, spectra were taken at points along the white line in Fig. 5a in steps of 33 nm. Figure 5b shows from top to bottom the spectra which were taken from left to right in Fig. 5a. It is clearly seen that the peak energy shifts to lower values when crossing a growth spiral center.

As seen in Figs. 4 and 5, for all samples the longest wavelength is always observed in the center of the spirals. This finding is similar to the results of Sugahara et al.¹⁰ and can be explained by a larger

Fig. 5. (a) Monochromatic CL image at 404.5 nm (3.07 eV) of the sample grown at 780°C; the white arrow indicates the line along which the spectra were taken. (b) CL spectra at points along the white line in (a) taken at 6 K.

QW thickness and/or higher indium content within the center of the spirals as compared with their peripheral regions. Also, some strain relaxation near the dislocations may occur. In principle, strain relaxation of a compressively strained QW leads to a redshift of the luminescence line. However, in the case of InGa_N QWs, a competing mechanism is the reduction of the piezoelectric field due to relaxation, resulting in a blueshift of the luminescence line. For a 3-nm-thick $\text{In}_{0.08}\text{Ga}_{0.92}\text{N}$ QW, if we assume a blueshift of 270 meV at full relaxation, using a strength of the piezoelectric field as in Ref. 15, this should overcompensate the redshift from strain relaxation of about 208 meV.¹⁶ Thus, the local strain relaxation near dislocations should not contribute significantly to the observed redshift of the emission wavelength inside the spirals.

First, we discuss the peak energy shift which could be caused by lateral thickness variations. Sonderegger et al.¹⁷ observed, for a single 1.5-nm-thick $\text{In}_{0.15}\text{GaN}$ QW, thickness variations of up to 1.5 nm, resulting in peak energy variations in the range of 100 meV, indicating a strong impact of thickness fluctuations on emission wavelength. Also, in our CL spectra we observe peak energy variations as large as 100 meV (Fig. 5b). From

theoretical calculations of the emission energy, assuming a constant lateral indium concentration of $x = 0.08$, such a shift corresponds to a thickness variation of 1.5 nm.¹⁸ Since in our MQW structure we have five QWs, one could assume a maximum lateral height difference of 7.5 nm, if the height differences were only due to QW thickness fluctuations and not to barrier thickness fluctuations. From our AFM measurements we observe the largest average height (8.6 nm) of growth spirals for the sample grown at the lowest temperature (760°C). In this case the observed lateral peak energy variation of 123 meV can be explained from QW thickness fluctuations alone. However, if only local thickness variations are assumed, the lateral peak energy fluctuations should become smaller with increasing growth temperature, since for the sample grown at the highest temperature (840°C) the average spiral height decreases to 3.7 nm. For this sample a lateral peak energy shift of 100 meV was observed. This implies additional sources of lateral peak energy shift.

One explanation could be indium accumulation in the center of the growth spirals, which was already found by Lu et al.¹² using backscattered electron images. This accumulation is more pronounced at higher temperature. This can be understood in terms of the faster surface diffusion of the indium atoms at higher T_G , which preferentially incorporate at the steps associated with the spirals. However, evidence for this assumption has to be provided by analytical transmission electron microscopy. For thick (80 nm) InGa_N layers, lateral indium concentration fluctuations have been observed already by Bertram et al.¹⁹ It should be noted that the randomly distributed dislocations are not connected with any wavelength shifts.

In addition to the wavelength distribution across the spiral domains, we also observe long-range wavelength fluctuations on a scale of several tens of microns at higher growth temperatures (820°C and 840°C). Moreover, surface features such as droplets and oval grooves were observed (arrow in Fig. 1b). SE imaging at higher magnification reveals that these grooves consist of closely spaced surface pits, as observed for the dislocation escape points. Therefore, one can assume that the oval area is surrounded by threading dislocations. We observed a significant and abrupt redshift of the peak wavelength and an increase of the FWHM inside such an oval groove.

In the AFM and SE images (Fig. 1), small randomly distributed surface pits appear, which we attribute to edge and mixed-type dislocations. Also, there are pits in the center of each growth spiral from screw dislocations. All these pits do not show any remarkable dark contrast in the CL images at 6 K, while the intensity distribution associated with the growth spirals dominates the CL contrast. Therefore, we conclude that the nonradiative recombination activity of dislocations, especially at

Fig. 6. (a) SE image and (b) monochromatic CL image at the low-energy slope taken at room temperature for the sample grown at 760°C.

6 K, is much smaller in InGaN MQWs than in GaN layers, where the dislocations appear as dark spots in the CL images (not shown here). From photoluminescence studies it is known that nonradiative recombination increases with higher temperature, either due to nonradiative recombination at defects such as dislocations and point defects or due to carrier escape from the QWs.²⁰ To study the recombination activity of dislocations at higher temperature we also took monochromatic CL images at room temperature. Figure 6 shows a monochromatic CL image of the InGaN QWs at the low-energy slope for the sample grown at 760°C. Again, we do not observe dark spots related to dislocations. Therefore, in our case, as proposed by Chichibu et al.,²¹ the increase in nonradiative recombination may mainly be caused by randomly distributed point defects and impurities. This finding is somewhat different from the results of Sugahara et al.,¹⁰ who found dark spots associated with dislocations in InGaN in CL images recorded at room temperature. This may be understood by different growth parameters, which could lead to a different number and distribution of point defects, which in their case may have accumulated at the dislocations.

CONCLUSIONS

We have studied the influence of growth temperature T_G (760°C to 840°C) on the optical and structural properties of $\text{In}_{0.08}\text{Ga}_{0.82}\text{N}/\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ MQWs emitting at 405 nm. We observed local emission line variations over growth spirals. The narrowest FWHM of the sum spectrum and thus the lowest wavelength variation was found for the sample grown at 780°C. The observed QW thickness fluctuations were highest for the sample grown at the lowest temperature (760°C). To

explain the observed local wavelength fluctuations, in addition to the thickness fluctuations, an inhomogeneous lateral indium distribution has to be assumed, with the highest indium concentration in the middle of the spiral domains. We did not observe dark spots solely related to nonradiative recombination at dislocations in the CL images of InGaN QWs. This could be one explanation for the fact that despite relatively high dislocation densities, LEDs and laser devices show high quantum efficiencies and high optical output powers. The AFM and CL results were used to optimize the growth conditions and hence the optical properties of MQWs serving as active layers in laser diodes.

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