Role of gross well-width fluctuations in bright, green-emitting single InGaN/GaN quantum well structures

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Gross well-width fluctuations have been observed by transmission electron microscopy (TEM) in single InGaN/GaN quantum wells (QWs) grown by metal-organic vapor phase epitaxy. Similar thickness variations are observed in commercial, green InGaN/GaN multi-QW light emitting diodes. Atomic force microscopy studies of equivalent epilayers suggest that these fluctuations arise from a network of interlinking InGaN strips, which are found (using TEM) to be indium rich at their centers. Plan-view TEM indicates that 90±8% of all threading dislocations (TDs) intersect the QW plane between the InGaN strips. Excitons may be localized at the strips' centers, preventing nonradiative recombination at TDs. © *2007 American Institute of Physics*.
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There exists a significant and continuing controversy about the mechanism of light emission in the InGaN quantum wells (QWs) which make up the active region of bright blue and green light emitting diodes (LEDs), given the high density of threading dislocations (TDs) found in working devices. In particular, various authors[1,2] have questioned the existence of nanometer-scale, indium-rich clusters, which had previously been postulated as localization centers within the QWs. In this context, a reassessment of the role of the broad-scale QW microstructure in controlling device performance seems apposite. For example, Hangleiter *et al.*[3] recently suggested a mechanism in which the microstructure (on a 100–200 nm scale) around the TDs gives rise to "self-screening" of the TDs. However, their work presents no evidence for the existence of the particular defect structure they suggest in current commercial devices.

Figure 1 shows a scanning transmission electron microscope high angle annular dark field (STEM-HAADF) image of the active region of a commercial green-emitting LED. The observed QWs exhibit gross fluctuations in width. Similar well-width fluctuations in LEDs have been reported by Narayan *et al.*[4], who suggested that these features might be instrumental in confining excitons away from TD cores. This suggestion assumes a "quantum disk" (QDisk) structure, similar to that suggested by Chichibu *et al.*[5], i.e., the existence of discrete InGaN disks in a GaN matrix. If such QDisks existed, with a density significantly in excess of the TD density, they would certainly help to prevent exciton diffusion to TDs. However, while the well-width variations seen in Fig. 1 are on a similar lateral scale (50–100 nm), to QDisks,[5] we show here that the structure of such QWs is not necessarily disklike. Nonetheless, such gross well-width fluctuations may play an important role in confining excitons away from TD cores.

We have examined InGaN epilayers and QWs grown by metal-organic vapor phase epitaxy on GaN pseudosubstrates[6] using a 6×2 in. Thomas Swan close-coupled showerhead reactor. 3.6 nm thick InGaN epilayers were grown, using trimethyl-indium, trimethyl-gallium, and ammonia as precursor gases with nitrogen as a carrier, and then annealed or heat treated as described later. For the quantum well structures, similar InGaN layers were capped with approximately 8 nm of GaN grown either at the InGaN growth temperature or at 860 °C. STEM-HAADF was performed on a FEI Tecnai F20 field emission gun TEM, coupled with an energy dispersive x-ray (EDX) detector for chemical analysis. Conventional TEM measurements were carried out on a JEOL 2000FX. Atomic force microscopy (AFM) was performed on a Veeco Dimension 3100 in tapping mode.

Well-width fluctuations are commonly seen in multi-QW (MQW) structures grown in our laboratory when a "two-temperature" (2T) method is employed.[7] Here we examine 2T single QWs: the InGaN was grown at 720 °C before the temperature was ramped to 860 °C for the growth of the subsequent GaN cap. During the temperature ramp no growth took place. Using cross-sectional STEM analysis we have observed that this 2T-QW [photoluminescence (PL) emission wavelength, λ=480 nm] exhibits gross well-width fluctuations similar to those observed in Fig. 1. Such fluctuations have also been observed, using STEM, in InGaN QWs grown at a single temperature (710 °C in this case) if the InGaN is annealed for 8 min at the growth temperature in an NH_3/N_2 atmosphere, prior to capping layer growth. A uniform QW structure, with no width fluctuations, was observed when the annealing step was not used. Both the annealing and 2T methods result in a blueshift and an increase in intensity of the room-temperature PL, as compared to an unan-

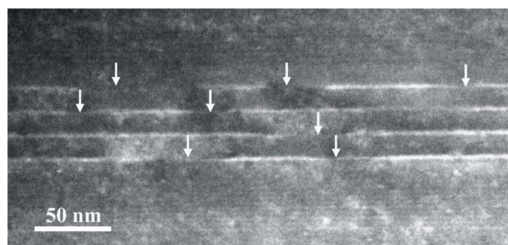

FIG. 1. Cross-section STEM-HAADF image of a commercial green LED showing gross thickness variations (arrowed) in all four InGaN QWs.

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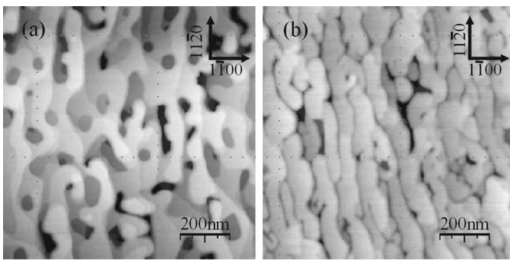

FIG. 2. AFM images of (a) a temperature-bounced InGaN epilayer and (b) an InGaN epilayer annealed at a single temperature; both show a network of interlinking InGaN strips, aligned roughly parallel to the $[11\bar{2}0]$ direction.

nealed, single temperature QW. To isolate the wavelength shift from any intensity rise, an annealed QW was grown at a reduced temperature, to achieve an emission wavelength similar to, or longer than, that of the unannealed QW. PL studies comparing this single temperature annealed QW ($\lambda = 515$ nm) and the unannealed QW ($\lambda = 507$ nm) reveal up to seven times greater peak emission for the annealed structure, despite the fact that it emits at a slightly longer wavelength.

To better understand the morphology corresponding to the well-width fluctuations observed in TEM, comparable epilayers were studied by AFM. To grow an InGaN epilayer roughly equivalent to the 2T-QW (i.e., without the GaN barrier but exposed to the higher temperature), we used a "temperature-bouncing" method: the InGaN epilayer was grown at 730 °C, subjected to a linear temperature ramp to 860 °C over 60 s, then cooled immediately. For the annealed epilayer, the temperature was reduced as quickly as possible after annealing for 8 min. The AFM data in Figs. 2(a) and 2(b) show that both the temperature-bounced and annealed epilayers consist of a network of interlinking InGaN strips, about 50–100 nm in width, aligned approximately parallel to the [11–20] direction. (In contrast, an unannealed epilayer presents a terraced morphology.[6]) For 2T methods, it is difficult to determine the actual temperature at which the observed changes occur and how well the temperature-bounced epilayer represents the 2T QW (since they do not undergo exactly the same thermal schedule). As a similar morphology may be observed due to both temperature bouncing and annealing, we have concentrated further TEM-based studies on annealed structures grown at a single temperature.

Figure 3(a) shows a STEM-HAADF image of a single InGaN strip in an annealed epilayer viewed down the [11-20] zone axis. The numbered crosses indicate the position of the probe for EDX analyses and the In:Ga ratio at each of these points is shown in Fig. 3(b). These data, and similar electron energy loss spectroscopy data for the annealed QW, suggest that the centers of the InGaN strips are indium rich with respect to their edges. Similar studies performed on an unannealed epilayer showed no evidence for varying indium concentrations.

Given this compositional nonuniformity, we postulate that excitons should be confined at the centers of the InGaN strips. However, for high emission efficiency, exciton localization needs to occur away from nonradiative recombination centers such as TDs. To determine if any relationship exists between the InGaN strips and the position of the TDs, TEM analysis was performed on the annealed epilayer in plan-view orientation. We used a bright-field multibeam imaging technique with the beam orientated along a {1-21-3} zone

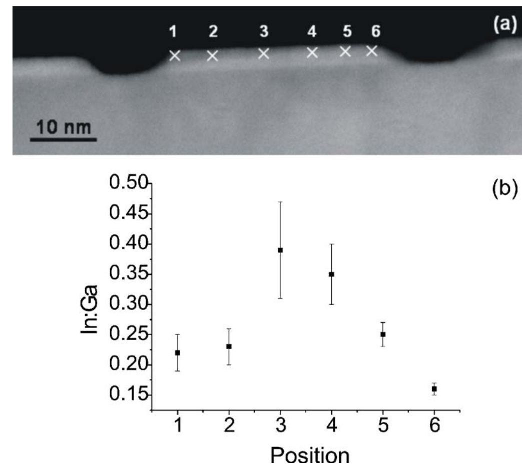

FIG. 3. (a) STEM-HAADF cross-section image of the annealed epilayer viewed along the $[11\bar{2}0]$ zone axis. The electron probe diameter was approximately 1 nm. The arrows indicate the probe positions for EDX analyses of the In:Ga ratio, as plotted in (b).

axis to reveal all types of dislocations;[8] the network structure may be observed simultaneously. Figure 4 shows a series of bright-field images taken down different {1-21-3} zone axes. Each of the images shows the same five TDs projected in different directions with the ends of the TDs that terminate at the network surface indicated by arrows. By comparing the three images, it is apparent that only one TD (circled) terminates at the center of an InGaN strip while the remaining four TDs terminate in, or very close to, the troughs between the strips. By analyzing several areas, we have determined that $90 \pm 8\%$ of the TDs terminate in the troughs between the interlinking InGaN strips. Thus, the gross well-width fluctuations do *not* correspond to QDisks, but instead to a network of interlinking InGaN strips with indium-rich centers. However, the majority of the TDs do not intersect these strips but instead pass through the GaN regions between them, leading to an increased band gap in the part of the active layer where the TDs are found, producing a self-screening effect not dissimilar to that proposed by Hangleiter *et al.*,[3] but related to a different microstructure.

Evidence from several techniques (PL, x-ray photoelectron spectroscopy, secondary ion mass spectroscopy, and TEM) shows that an overall decrease in the average indium content of the epilayer occurs during the annealing process. It has also been observed by TEM that the epilayer thickness does not measurably change on annealing, except at the strip edges and between the strips. Thus, the observed troughs may form by decomposition of indium-rich regions. Srinivasan *et al.*[9] have demonstrated using cathodoluminescence that indium tends to segregate to TD cores, for 100 nm thick InGaN layers, and given our proposition that troughs form

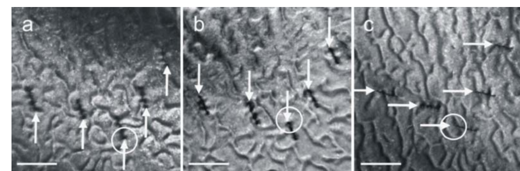

FIG. 4. Bright-field TEM images of an annealed InGaN epilayer taken on the $\{1\bar{2}1\bar{3}\}$ family of zone axes showing five TDs projected in three different directions. The arrows indicate where the TDs terminate at the surface. Scale bar=200 nm.

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by decomposition of indium-rich regions, these data may explain why the dislocations are found between the InGaN strips in the annealed epilayer. We note that this mechanism does not fully explain the existence of indium-rich regions at the centers of the InGaN strips.

Previous PL studies suggest a lateral length scale of localization of approximately 2 nm for QWs grown at Cambridge. In the absence of nanometer-scale In-rich clusters, this may be attributed to monolayer variations in QW thickness at this length scale.[10] However, more than one mechanism of exciton localization may be active, with different length scales being relevant to different mechanisms. At room temperature or above, carriers may hop between nanometer-scale localization sites due to, e.g., thermionic emission, but may still be prevented from reaching dislocation cores by the broader microstructure described here.

In conclusion, using AFM and TEM we have shown that the actual structure of single InGaN QWs with gross well-width fluctuations is comprised of interlinking strips of InGaN, whose centers are indium rich, compared to their edges. We postulate that excitons are localized at these indium-rich regions preventing them reaching TDs, which predominantly intersect the QW plane near the edges of, or between, the InGaN strips. The microstructures of our QWs are similar to those observed in cross-section studies of MQWs in commercial devices and hence these data may explain the performance of bright commercial green LEDs.

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