In$_{x}$Ga$_{1-x}$/GaN quantum wells (QWs) are used as active region in light-emitting diodes (LEDs) and laser diodes. By varying the In concentration, one is theoretically able to tune the peak emission between infrared and ultraviolet.\footnote{Present address: SIDMAR, Belgium.} A typical device has, starting from the substrate, a n-i-p structure with the n-doped region next to the substrate. The intrinsic part is the QW structure. n-type doped GaN can be rather easily obtained with Si as a donor.\footnote{Author to whom correspondence should be addressed; electronic mail: benny.vandaele@ua.ac.be} The p-type region however, is much harder to grow. In general, Mg atoms are used as acceptors. The drawback is that the Mg atoms may react and form Mg–H complexes;\footnote{Received 1 June 2004; accepted 8 September 2004} hydrogen being present in high amounts in the metallocarbide vapor phase epitaxy (MOVPE) reactors [ammonia (NH$_3$), trimethylgallium (TMGa), and carrier gas]. These complexes are electrically inactive. However, the hydrogen can be removed by low-energy electron irradiation\footnote{M. R. Leys IMEC-Department of Information Technology (INTEC), University of Ghent, Belgium} or by thermal annealing at temperatures above 600 °C.\footnote{B. Van Daele a and I. Moerman IMEC-Department of Information Technology (INTEC), University of Ghent, Belgium} As the Mg precursor has an undesirable memory effect on the subsequently grown layers, we adopted the policy to first grow a set of multiquantum-well (MQW) structures under different growth conditions without a p-doped top layer. In a second step, these structures were regrown with Mg-doped GaN at a temperature of 1040 °C. It is an accepted fact that high-quality GaN requires high growth temperatures, which is in strong contrast to the much lower optimal growth temperature for the InGaN/GaN active region. The lower growth temperatures for the InGaN are necessary because InN already decomposes in a vacuum at 630 °C.\footnote{K. Jacobs b and G. Van Tendeloo IMEC, Belgium} The use of high temperatures for the p-type capping layer has the advantage that it results in a perfect crystalline structure. No pyramidal defects or other kinds of Mg clustering, as the ones reported in p-type layers grown at 950 °C,\footnote{© 2004 American Institute of Physics} are observed. However, there is one major drawback: Above a certain nominal In concentration, the capping of the active region turns the wafers black purple. Comparison with a noncapped part shows that the originally optically active sample loses, all room-temperature photoluminescence (RT-PL) signals. In high-resolution x-ray diffraction (HR-XRD), the contribution of the InGaN layers vanishes; no satellite reflections related to the artificial superstructure can be retrieved,\footnote{In$_{x}$Ga$_{1-x}$/N/GaN MQWs, consisting of 5 In$_{x}$Ga$_{1-x}$/N QWs, have been grown on (0001) sapphire in a 3 × 2 in. close coupled showerhead MOVPE reactor. NH$_3$, TMGa, and trimethylindium (TMIn) were used as precursors, N$_2$ was used as the carrier gas. The general sample structure consists of a 25 nm thick low-temperature buffer layer, 1.4 μm undoped GaN, 1.5–2.0 μm GaN:Si (both grown at 1060 °C), the active region and, in some samples, a GaN:Mg cap layer (grown at 1040 °C). The active region of the samples studied were all grown with a TMIn/(TMIn + TMGa) ratio of 0.5, the temperature (the same for the InGaN QWs and the GaN barriers) was varied from 660 °C to 680 °C and the pressure was 300 Torr. In a few samples, growth interruptions of 5 and 10 s after InGaN growth have been used. InGaN layers are typically 4–5 nm thick, GaN buffer layers 8–10 nm. Samples have been characterized by TEM techniques, HR-XRD, and RT-PL. The local In concentration, determined by TEM, has been calculated starting from the local lattice strain using the linear elasticity theory and assuming that Végard’s law is valid for In$_{x}$Ga$_{1-x}$/N. The lattice parameters and elastic coefficients, for GaN and InN, have been taken from Refs. 9 and 10, respectively. The large error bars obtained with this technique are due to the fact that the relaxation in TEM samples, as thin as a few tens of nanometers, is unknown. The lattice strain has been evaluated with the DALI software package.\footnote{Electron-beam irradiation at 200 kV inside the electron microscope is known to introduce clustering in the InGaN after a few minutes.\footnote{Samples grown at 660 °C, without growth interruption, systematically turn black purple during/after the p-type capping. The same effect occurs during thermal annealing in a N$_2$ or NH$_3$ ambient, indicating that the raise in temperature is} This effect has also been observed in our samples. Therefore, special care has been taken in order to work with minimum exposure. The clustering reported here is therefore intrinsic, not induced by the electron beam.} Samples grown at 660 °C, without growth interruption, systematically turn black purple during/after the p-type capping. The same effect occurs during thermal annealing in a N$_2$ or NH$_3$ ambient, indicating that the raise in temperature is
InGaN / GaN MQW region has transformed in a GaN matrix to fluctuate between 15% and 30%. After capping, the concentration in the surrounding matrix has been determined.

Metallic In thickness. High-resolution TEM reveals the structure of metallic In platelets in the GaN matrix. These platelets and the formation of the dislocations. The general properties of the metallic In platelets will be described below. The platelets, studied by high-resolution TEM, have a fixed orientation relation with the GaN matrix: (0001)$_{\text{GaN}}$// (101)$_{\text{In}}$ and [1120]$_{\text{GaN}}$// [111]$_{\text{In}}$ (Fig. 2). This is not surprising since the In (101) planes have a pseudohexagonal atom configuration. The In/GaN interface is sharp. For counting the TD density in these samples, avoiding possible blocking and/or bending by the MQW structure, we decided to remove the MQW region by ion milling. This was not directly successful: In some of the samples grown at 680 °C, we ended up with a plan-view specimen somewhere in the QW region. The plan-view specimens revealed the In platelets and show dislocation networks (and few other defects) in the direct neighborhood of the platelets (Fig. 3). These dislocations, lying in the GaN (0001) planes, all have as a Burger’s vector 1/3[1120] (or equivalent by symmetry). In cross section specimens, the dislocations can be found near the platelets at the same height, clearly indicating a connection between the diffusion of the In into the metallic platelets and the formation of the dislocations.
The decomposition of InGaN into GaN and metallic In clearly is a thermally activated first-order process. The interface between GaN and In is sharp; no In-rich InGaN is found at the edges. Intermediate structures, such as In-rich InGaN regions the size of the In platelets, have not been found either. The presence of dislocations indicates diffusion of In and Ga. The question of how the metallization process has been triggered is more difficult to answer. The segregation process only occurs within the QWs. Only occasionally are the In platelets larger than the original QWs, proving a low solubility of In in GaN at the growth temperature of the p-type GaN. The possible presence of a liquid In_{x}Ga_{1−x} alloy has been predicted theoretically. According to these calculations, the liquid phase can already be formed at a temperature above 750 °C, indicating that the QW region should not be heated at all. However, under some conditions, the formation of the metallic In platelets can be restricted. The anomaly using the growth interruption indicates that initiation centers are important; the metallic In needs a seed or nucleus to start growing. These nuclei are removed by the growth interruption. It is important to note that the total amount of In in the QWs grown at 660 °C is quite high. This can be seen by comparing the volume of the In platelets with the volume of the GaN in the former QWs (Fig. 1). The samples grown at 680 °C have a lower average In concentration and show a much smaller density of In platelets. Therefore, the initiation centers are more likely formed with a higher In concentration in the InGaN. However, the total amount of In, present in the QWs, is not a good measure of the formation probability. This is shown by the fact that the In platelets have not been observed in the samples grown at 660 °C with growth interruption.

Considering all experimental information, it is most likely that the initiation centers are some small In droplets. They can be avoided by growing in InN evaporating conditions (high growth temperature and use of a growth interruption). The fact that the growth interruption only affects the top part of the QWs indicates that the proposed In droplets are located at the surface of the QWs. This points to the presence of an In floating layer during the growth of InGaN, in analogy with the presence of an In floating during the growth of InGaAs. Assuming the presence of an In floating layer during the InGaN growth explains all observations. At a higher growth temperature, there is more In evaporation out of the floating layer, resulting in a lower In content in the InGaN QWs and fewer initiation centers at the surface of the QWs. Growth interruption removes the top of the QWs and therefore also the In floating layer. Consequently, the initiation centers for metallic In formation are then also removed.

This work has been performed within the framework of IAP V-1 and was supported by the Institute for the Promotion of Innovation by Science and Technology in Flanders (IWT-Project No. 980319). Two of the authors (B.v.D. and K.J.) are grateful to the Fund for Scientific Research—Flanders (F.W.O.—Vlaanderen), Belgium.

13This value may be even higher. The dimensions of the QDs are much smaller than the typical thickness of a TEM specimen. In reality, one measures the concentration averaged along the electron-beam direction.