## 2.7 Indium segregation in (Ga,In)(N,As) multiple quantum wells

$Ga_{1-x}In_xN_yAs_{1-y}$ multiple quantum wells (MQWs) with $x > 0.35$ and $y > 0.04$ can be grown by molecular-beam epitaxy (MBE) under growth conditions close to stoichiometry in combination with substrate temperatures as low as 350 °C. Despite the high photoluminescence efficiency reported for these QWs, a clear understanding of the effect of the low substrate temperature ($T_s$) and the V/III ratio on the growth kinetics and on the morphological, structural, and compositional properties is still missing. In general, independent of the growth conditions, the indium segregation in the quaternary alloy is so far a widely unexplored subject.

$In_{0.36}Ga_{0.64}N_{0.04}As_{0.96}/GaN_{0.008}As_{0.992}$ MQWs with a well thickness of 4 nm and a barrier thickness of 11 nm were analyzed by transmission electron microscopy (TEM). With the combined analysis of the (002) dark-field (DF) and high-resolution TEM images, we determined the local In and N concentration profiles along the growth direction. The MQWs were grown on GaAs(001) substrates by plasma-assisted MBE. For $T_s = 375$ °C, we found that the modification of the V/III ratio, which is determined from the beam equivalent pressure (BEP) of the elements as $R_{\mathrm{BEP}} = (\mathrm{BEP}_{\mathrm{As}})/(\mathrm{BEP}_{\mathrm{Ga}} + \mathrm{BEP}_{\mathrm{In}})$, from 3 to 50 has a critical effect on the indium segregation.

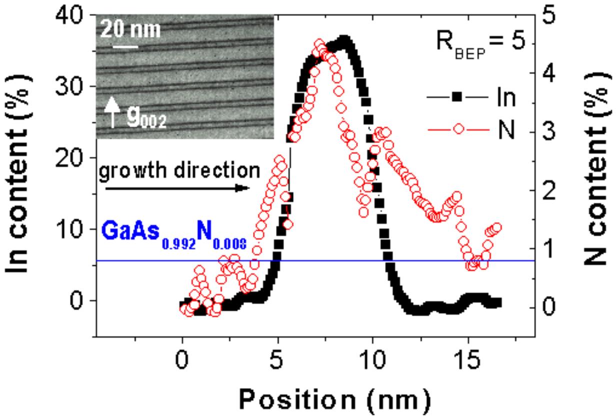

Fig. 18. In and N concentration profiles of (Ga,In)(N,As) MQWs grown at $T_s = 375$ °C and $R_{\mathrm{BEP}} = 5$. Inset: DF-TEM image.

Figure 18 shows the In and N distribution profiles for the QWs grown at $T_s = 375$ °C and $R_{\mathrm{BEP}} = 5$. Independent of the $R_{\mathrm{BEP}}$ used, we obtained morphologically perfect two-dimensional QWs showing laterally homogeneous composition (cf. inset of Fig. 18). Concerning the element distribution inside the QW, the indium profile reveals a pronounced asymmetry, characteristic of segregation. The nitrogen profile exhibits a strong asymmetry as well. As observed, areas of lower N content correspond to areas of higher In content, a well-known result because of the preferred formation of Ga-N and In-As bond configurations. As shown in Fig. 19(a), increasing the V/III ratio ($R_{\mathrm{BEP}} = 50$) reduces the In segregation resulting in a more symmetric In profile. We found that the impact of $T_s$ on the In segregation is masked by the increasing surface roughening during growth and the appearance of lateral composition fluctuations.

Segregation is normally described and quantified using the segregation efficiency, which defines, in the notation of K. Muraki *et al.* [Appl. Phys. Lett. **61**, 557 (1992)], the fraction of In atoms on the topmost layer that move to the next layer. As shown in Fig. 19(b), the asymmetric In distribution inside the QW of Fig. 18 can be very well described by Muraki's model,

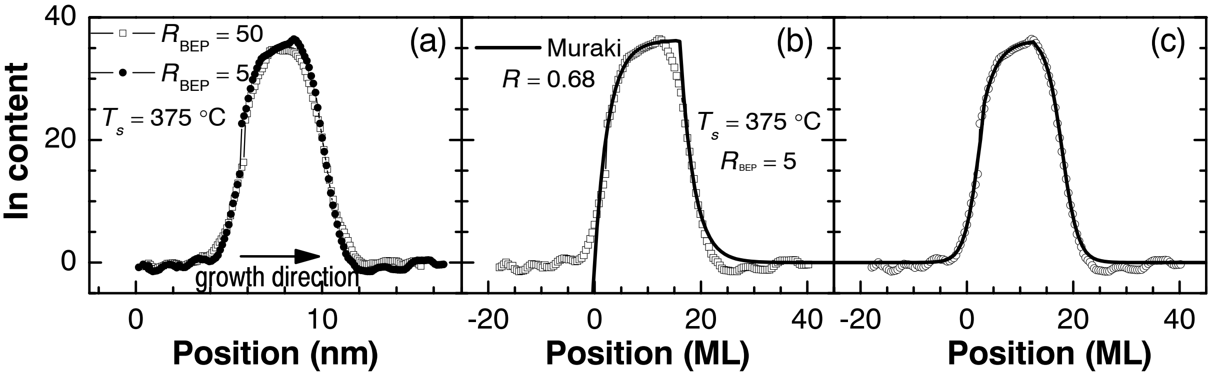

Fig. 19. (a) Increasing the V/III ratio reduces the In segregation. (b) Fit to Muraki's model for segregation. The disagreement at the second interface is apparent. (c) Perfect description on the In distribution using our extension of Muraki's model to include growth-related diffusion processes at the interfaces.

extracting an efficiency of $R = 0.68$, i.e., the In segregation is low. However, the deviation of the fit to the data close to the second interface is apparent. In particular, Muraki's model predicts a higher In content at the interface than experimentally observed. The comparison to other segregation models gives a similar disagreement. We therefore attribute this deficiency of the segregation models to an incorrect description of the interfaces of the (Ga,In)(N,As) QWs. A different behavior of the In atoms in the center of the QW compared to the ones at the interfaces explains these discrepancies: the In distribution inside the QW is governed by *segregation*, whereas the In distribution at the interfaces is mainly determined by a growth-related *diffusion* process, affected by the phase decomposition tendency.

We found that the concentration profiles at the interfaces can be very well described using a phenomenological expression based on a double sigmoidal function with the interface thickness $L$ as the main fitting parameter. As shown in Fig. 19(c), the combination of this approach with Muraki's model results in a perfect description of the complete indium profile in (Ga,In)(N,As) QWs. This extends Muraki's model for segregation by taking into account the interfaces. Note that in thin QWs (4 nm), any contribution coming from the interfaces (about 0.5 nm) is significant because of the comparable length scales. For the QWs discussed here, we found $L = 1.5$ monolayers (ML) corresponding to rather abrupt interfaces. QWs grown at $T_s = 390$ °C and $R_{\mathrm{BEP}} = 5$ are characterized by $L = 2.9$ ML. This large interface broadening cannot be explained by the enhanced, thermally activated diffusion at the interfaces due to the increase of $T_s$ alone. This would produce a broadening of only 0.4 ML so that the existence of an additional strongly $T_s$ dependent mechanism is suggested. We know that the increase of $T_s$ favors the formation of In-As bonds. Therefore, the permanent As supply during growth will extend the In distribution into a broader interfacial width as was indeed observed experimentally. We found that diffusion at the interfaces of (Ga,In)(N,As) QWs is triggered by the inherent phase separation tendency of the alloy.

(E. Luna, F. Ishikawa, Á. Guzmán, A. Trampert, K. H. Ploog)