Structural defects characterisation of GaInNAs MQWs by TEM and PL

M. Herrera, D. González, R. García, M. Hopkinson, P. Navaretti, M. Gutiérrez and H.Y. Liu

Abstract: The authors' work is focused on the structural and optical properties of GaInNAs/GaAs(001) QWs with In and N contents in the ranges 0.20–0.35 and 0.013–0.023, respectively, studied by transmission electron microscopy and photoluminescence. An undulation of the wells was observed when increasing the In content to 25% and the N content to 1.6%. This induces the appearance of extrinsic Frank dislocation loops, which are most likely formed because of local stress accumulation in the structure. The PL spectra show intense peaks at 1.1 μm and 1.2 μm for samples with In compositions of 20% and 25%, respectively, representing therefore structures with and without loops. Consequently, the existence of these structural defects seems not to affect the optical efficiency of this alloy. On increasing the In content to 35%, a considerable decrease in the PL peak intensity, related to the existence of threading dislocations observed by TEM, has been found. The unfaulting of dislocation loops by means of the reaction with two Shockley partial dislocations is proposed as the mechanism of formation of these threading dislocations.

1 Introduction

Recently, there has been a great deal of work done on the $GaInN_xAs_{1-x}$ ($x < 0.05$) material system. The application of this alloy for light sources in the fibre optics communications range (1.3–1.5 μm) appears to be a promising alternative to the classical GaInAsP/InP system. The GaInNAs alloy offers the advantage of better electron confinement in the active region due to a higher conduction band offset when combined with GaAs barriers [1], which leads to enhanced high-temperature device performance. Furthermore, GaInNAs shows an extremely large negative bandgap bowing [2] that allows a red shift of the emission wavelength by the addition of relatively low (<5%) N contents to GaInAs structures. The small atomic size of N atoms reduces the lattice mismatch with GaAs substrates and, consequently, less structural defects are expected. However, it has been reported that a degradation of the optical properties of GaInNAs structures occurs on increasing the N content [3], showing lower PL intensities and wider linewidths. This has been associated with composition fluctuations and clustering, resulting in nonradiative centres. Therefore, typical laser results are reported for devices with a nitrogen content of around 1% and with the In concentration maximized [4, 5]. Nevertheless, the appearance of structural defects is more likely for structures with these high In concentrations, due to the higher strain. We have studied the influence of increasing In and N compositions on the structural and optical properties of GaInNAs multiquantum wells.

2 Experimental

GaInNAs multiquantum well structures with different In and N contents were grown on (001) on-axis GaAs substrates in a VG V80H molecular beam epitaxy (MBE) system equipped with an Oxford Applied Research HD25 radiofrequency plasma source for N. The N composition was controlled by monitoring the intensity of the atomic N plasma emission and calibrated from the X-ray diffraction analysis of bulk samples grown under similar conditions. The samples studied consist of a stack of five GaInNAs quantum wells grown at 460°C and sandwiched between $GaAsN_{0.007}$ barriers. The composition of the quantum wells (QWs) in each of the three samples considered is as follows: sample A20, 20% In and 1.3% N; A25, 25% In and 1.6% N; A35, 35% In and 2.3% N.

Samples were prepared for transmission electron microscopy (TEM) by mechanical thinning followed by ion milling for cross-section observation (XTEM). The TEM study was performed using a JEOL 1200EX transmission electron microscope operating at 120 kV, using mainly the 220BF and 002DF reflections. The structures were also studied by photoluminescence (PL) using He–Ne laser excitation and an InGaAs detector.

3 Results

Figure 1*a* shows a 002DF transmission electron microscopy (TEM) micrograph of the sample with 20% In (A20), exhibiting flat GaInNAs QWs, with no structural defects. When increasing the In content to 25%, however, periodic modulations of the upper surface of the wells are developed, with period of 20 nm, as revealed in Fig. 1*b*. Moreover, Frank dislocation loops with size of approximately 20–30 nm have been observed in all the wells. These

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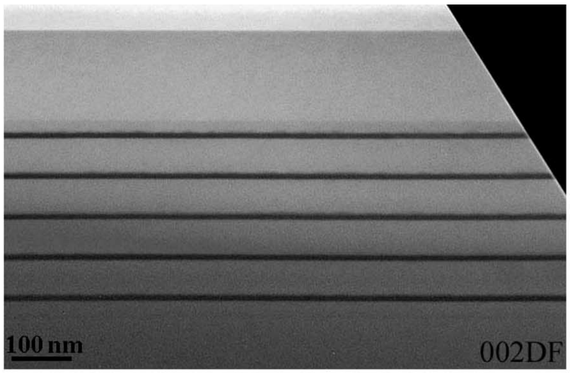

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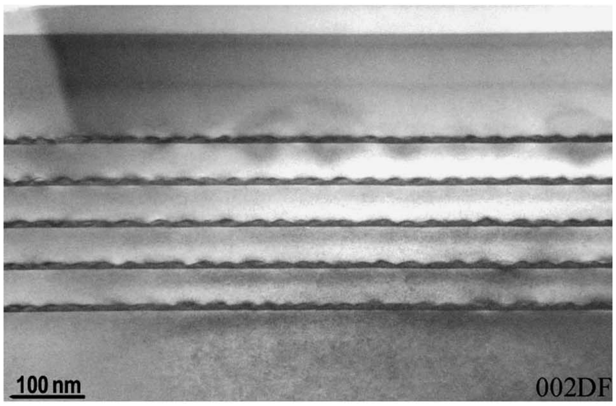

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Fig. 1 *002DF XTEM images*

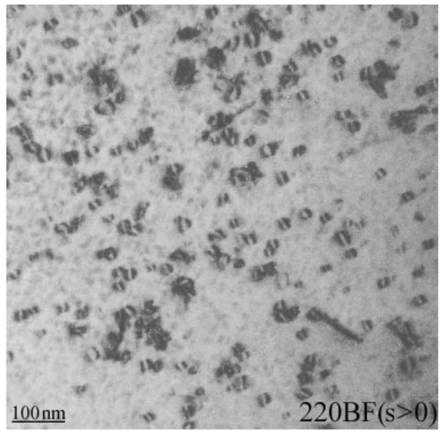
a $Ga_{0.8}In_{0.2}N_{0.013}As$ quantum wells, perfectly flat
b $Ga_{0.75}In_{0.25}N_{0.016}As$ quantum wells, undulated with period 20 nm approximately

loops have been characterised by the inside–outside contrast criterion as extrinsic, i.e. with an extra layer present with regard to the perfect arrangement of the crystal.

The sample with higher In and N compositions in our work, A35, has also shown the undulation of the GaInNAs QWs, as well as the appearance of extrinsic Frank dislocation loops, as can be observed in the plan view (PV)-TEM micrograph of Fig. 2*a*. In addition, threading dislocations (TDs) has been found in this structure, with a nonuniform density, on average $3–4 \times 10^9\,cm^{-2}$ (Fig. 2*b*). The characterisation of the threading dislocations by means of the invisibility criterion has given the result that they have Burger vectors of $1/2a\langle 110\rangle$ type.

However, whereas approximately two thirds of the TDs have the Burgers vector out of the growth plane (60°-out), one third have the Burgers vector parallel to the growth plane (60°-in), as follows: 35% of the Burgers vector is $1/2a\langle 110]$, 27% approximately is $1/2a\langle 101]$ and the remaining 38% is $1/2a\langle 011]$. We note that there are no conventional misfit dislocations in any of the QWs.

The PL spectra at room temperature of the GaInNAs structures are shown in Fig. 3. The emission wavelength of sample A20 is approximately 1.1 μm, with an intense, narrow peak. A high PL efficiency is also obtained when increasing the In content to 25% (A25), and the position of the peak is shifted to 1.2 μm. For the structure with 35% In composition (sample A35), an emission wavelength of 1.3 μm approximately is reached. Nevertheless, the emission efficiency is highly degraded, probably due to the structural defects observed in the TEM analysis.



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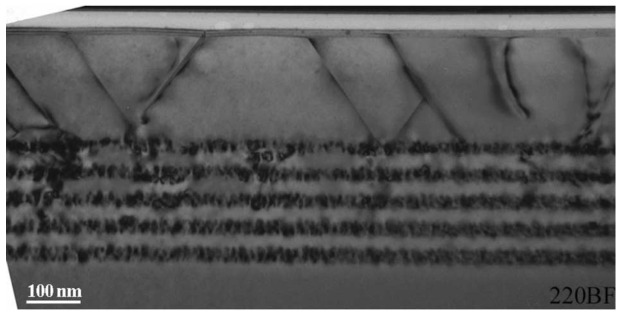

b

Fig. 2 *220BF images of sample A35 ($Ga_{0.65}In_{0.35}N_{0.023}As$ quantum wells)*

a PVTEM micrograph with $s > 0$, showing a high density of Frank dislocation loops
b XTEM micrograph, where threading dislocations are observed

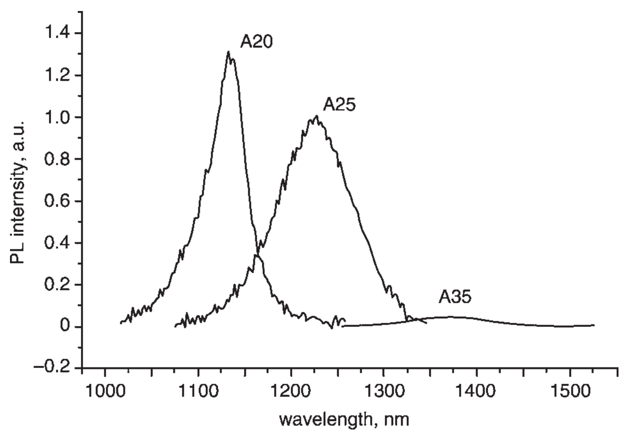

Fig. 3 *PL spectra of the studied samples*

Intense and narrow peaks are observed for $Ga_{0.8}In_{0.2}N_{0.013}As$ and $Ga_{0.75}In_{0.25}N_{0.016}As$ quantum wells (samples A20 and A25), while the PL efficiency is degraded for the $Ga_{0.65}In_{0.35}N_{0.023}As$ structure (sample A35)

4 Discussion

Characterisation by transmission electron microscopy of GaInNAs structures has shown that an undulation of the QWs is induced when increasing the In content from 20% to 25%. This is the classical behaviour of high In content GaInAs structures [6]. However, this change in surface morphology has been reported to take place at lower In compositions in the dilute nitrides than in GaInAs because of a phase separation into In-rich and In-deficient regions [7]. Our results have revealed that in the case of GaInNAs structures, the undulation of the QWs is accompanied by the formation of small Frank dislocation loops. We believe that the appearance of these loops is directly linked to the 3-D morphology. Dislocation loops are usually observed in structures with high local stress concentrations, as in the growth of quantum dots (QDs), but they have not been found in GaInAs QWs. Thus, the cause for its appearance in GaInNAs should be related to the incorporation of N in the material. Extrinsic Frank dislocation loops are constituted by a double stacking fault sequence. It is possible that the addition of small amounts of nitrogen to the GaInAs alloy change significantly the stacking fault energy (SFE) of the material. The influence of N on the stacking fault energy has been calculated for metals such as FCC iron-based alloys (with a nitrogen content lower that 0.5%) [8], where it is predicted first there is an increase of the SFE for low N content and then a rapid decrease of the SFE. We have not found theoretical or experimental predictions for the influence of N on the SFE for the GaInNAs alloy, but our results suggest a decrease of the stacking fault energy by the addition of 2% N in GaInAs structures. This fact, together with the high level of local stresses in the undulated GaInNAs QWs could favour the generation of Frank loops in our structures.

The next stage in the formation of structural defects in the studied GaInNAs quantum wells has been found when increasing the In content to 35%. Threading dislocations with a nonuniform distribution and with a Burgers vector characterised as $1/2a\langle 110\rangle$ are observed. However, as discussed previously, whereas two thirds approximately of the TDs have the Burgers vector out of the growth plane ($1/2a[101]$ and $1/2a[01\bar{1}]$), one third possess Burgers vectors parallel to the growth plane, $1/2a[110]$. This is an unexpected result, given that in GaAs(001)-based structures classical dislocations usually have the Burgers vector 60°-out. The formation of the first 60°-out dislocations in epitaxial layers is usually attributed to the Mathews and Blakeslee model [9], but in our case the thickness of the quantum wells is lower than the critical layer thickness predicted by this model. However, in nonplanar surfaces as in quantum dots or undulated quantum wells, it has been reported that concentration of stresses in the compression areas between the dots could produce the formation of small fragments of misfit dislocations that would later glide towards the surface, giving rise to the threading dislocations [10]. However, misfit dislocations have not been observed in the TEM study of this high In content structure. Furthermore, the particular form of these dislocations, the 60°-in type, could never be formed from misfit dislocations that glide towards the surface, since misfit dislocations with this Burgers vector would be sessile. Therefore, the origin of all the observed TDs (both with the 60°-in and 60°-out Burgers vector) needs to be clarified. We suggest a common origin for all the TDs in the form of an unfaulting of the existing Frank loops into perfect dislocation loops and their subsequent glide movement towards the sample surface. It has been widely reported that an extrinsic Frank loop can be unfaulted into a perfect one by the coalescence reaction with two Shockley partials [11, 12]. There are three Shockley partials parallel to each $\{111\}$ habitual plane of a Frank loop, which is the prerequisite for the unfaulting process. The reaction of each of the three available pairs of Shockley partials with an imperfect loop would be the source of dislocations with three different Burgers vector of type $1/2a\langle 110\rangle$. Two of them would be 60°-out and the third one would be 60°-in. This result is in good agreement with our experimental observations of the amount and type of characterised Burgers vectors. Therefore, we believe that the origin of the studied distribution of TDs is the mechanism of unfaulting of pre-existing loops.

With regard to the PL results, the photoluminescence spectra for sample A20 (composition of 20% In and 1.3% N) shows an intense PL peak at 1.1 μm. This wavelength is red-shifted to 1.2 μm on increasing the In and N contents to 25% and 1.6% (sample A25), respectively, as expected. For an In composition of 35% (A35), the PL peak reaches 1.3 μm, but its intensity falls considerably. The most obvious structural cause of this behaviour is the threading dislocations observed in this sample by TEM, but it is worth noting the high emission efficiency of sample A25, in spite of the high density of dislocation loops in this structure. There would appear to be no direct influence of dislocation loops on the optoelectronic behaviour of this material. The dislocation loops appear to be somewhat benign and qualitatively different from the effect of threading dislocations. This result is in contrast to the results obtained by Volovik *et al.* [7], who attributed the degradation of the luminescence intensity in GaInNAs samples to the appearance of dislocation loops. The influence of dislocation loops in the PL emission has been investigated for silicon structures where, paradoxically, the formation of these loops has been used as a tool to enhance the radiative emission in this material by encouraging localisation. It is reported that dislocation loops introduce a localised strain field that modifies the bandgap of the structure, providing spatial confinement in three dimensions. If, in the GaInAsN material, capture is encouraged in the vicinity of dislocation loops, we must conclude that the loops themselves do not act as significant nonradiative recombination centres in these materials.

5 Conclusions

We have investigated the effect of increasing the In and N compositions on the structural and optical properties of GaInNAs QWs. Our results have shown the undulation of the wells and the formation of extrinsic Frank dislocation loops when increasing the In content from 20 to 25%. The appearance of these defects could be the result of higher local stress concentrations in the material because of the introduction of N and the 3-D morphology. Good PL characteristics have been observed for samples with 20 and 25% In content, at wavelengths 1.1 and 1.2 μm, respectively. This means that the presence of Frank dislocation loops does not degrade the optical quality of these structures. For higher In contents (35%), threading dislocations appear and there is a rapid quenching of the photoluminescence efficiency. The unfaulting of dislocation loops occurring as a consequence of a coalesce reaction between two Shockley partials is proposed as a mechanism for the formation of the observed threading dislocations.

6 Acknowledgments

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