

Structural and optical properties of pseudomorphic $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys

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Thick (225 nm) $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers, grown on 5 μm thick GaN, were found by x-ray diffraction (XRD) measurements to be pseudomorphic up to $x=0.114$. Transmission electron microscopy showed that no misfit or additional threading dislocations were created at the $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ interface. Composition of the overlayers was determined by Rutherford backscattering spectrometry and correlated to both the a and c lattice constants from XRD. It was found that Vegard's law is applicable at these compositions, if the biaxial strain is included. Biaxial strain must also be considered to accurately determine the bowing parameter as shown by optical transmission measurements. © 1998 American Institute of Physics. [S0003-6951(98)00539-7]

The band gaps of $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys cover a wide spectral range, from red (InN) to UV (GaN), making this alloy system ideal for optoelectronic applications as light-emitting and laser diodes.^{1,2} The relationship between the emission wavelength with the In composition has usually been correlated to the band-gap energy with a bowing parameter of 1 eV.^{3,4} This has arisen since the composition of the $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers is typically determined from the c lattice parameter, measured by x-ray diffraction (XRD). This assumes that the layers are relaxed and follow Vegard's law.⁵

In this letter we report that $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers grown on GaN are in biaxial compression. Therefore, to determine the composition by XRD, both the c and a lattice parameters must be measured, and the elastic constants of $\text{In}_x\text{Ga}_{1-x}\text{N}$ must be known. We have previously reported that $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers grown on GaN on sapphire substrates are pseudomorphic for thickness $t=225$ nm at x up to 0.114.⁶ This is well beyond the critical thickness typical for other III-V semiconductor alloys,⁷ and furthermore, the In composition exceeds the equilibrium solid solubility limit for indium in GaN determined by Ho and Stringfellow.⁸ More recently, pseudomorphic growth has also been found for $x < 0.20$ at $t=40$ nm.⁹ In the latter work, the elastic constants used to determine the composition were interpolated from published values for InN and GaN.⁸ However, it is worth noting the reported values for the elastic constants vary over a large range.¹⁰

In this work, we provide additional evidence that thick (225 nm) $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers grown on GaN are pseudomorphic to the underlying GaN layer up to $x=0.114$ without the presence of misfit dislocations at the $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ interface. The composition of the $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers was measured by Rutherford backscattering spectrometry (RBS) and correlated to high-resolution x-ray diffraction (HRXRD) measurements of the a and c lattice constants. In addition, the energy-band gap of the strained alloys was measured and compared to the composition. We further make the compari-

son to the composition and optical properties reported in the literature for thin $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers used in device structures by assuming they are also pseudomorphic.⁴ This results in a larger bowing parameter than previously reported for $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys.⁴

$\text{In}_x\text{Ga}_{1-x}\text{N}$ layers (225 nm thick) were grown at 750 °C on 5 μm thick GaN on c -plane sapphire by metalorganic chemical vapor deposition (MOCVD). The a and c lattice constants were determined by measuring the (10 $\bar{1}2$) and (0002) reflections, respectively, using a high-resolution triple-axis diffractometer with Cu $K\alpha$ radiation. Compositions were determined from RBS by using 2 MeV He ions with an uncertainty in $x < 0.01$. The film thickness and defect structure were analyzed with cross-section transmission electron microscopy (XTEM). The XTEM was carried out at 300 kV on samples that were mechanically thinned to < 5 μm and ion milled to electron transparency using a liquid-nitrogen cold stage. Chemical x-ray (EDX) analysis was carried out in the TEM to confirm the location of the $\text{In}_x\text{Ga}_{1-x}\text{N}$ layer. The band gap of the $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers was determined by optical transmission.

The maximum In composition to obtain single-phase $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys was determined by RBS to be $x=0.114$. The full width half maximum $\Delta\omega$ of the (0002) reflection was found to increase with x (see Table I). For $x > 0.114$, the composition of the films was inhomogeneous as indicated by RBS and XRD measurements. In these films, several peaks were found in the XRD, corresponding to different compo-

TABLE I. Values for the composition x of $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys determined by RBS and calculated from XRD measurements. The full width half maximum, ω , of the rocking curve is from XRD in units of minutes. Calculated values of the critical thickness and the strain are also tabulated for each of the alloy compositions.

x (RBS)	x , pseudo	x , (0002)	Crit t , nm	Strain, %	$\Delta\omega$ (0002), min
0.054	0.055	0.076	12.8	0.21	6.70
0.072	0.072	0.104	8.7	0.32	8.80
0.100	0.097	0.142	5.5	0.37	12.90
0.114	0.115	0.167	4.6	0.51	19.30

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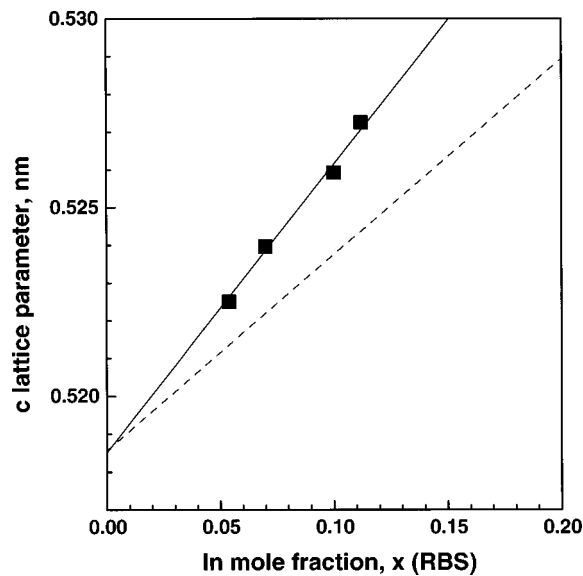


FIG. 1. XRD measurement of the c lattice parameter versus composition x of $\text{In}_x\text{Ga}_{1-x}\text{N}$ determined by RBS. The solid line is a least-squares linear fit of the data. The dashed line is a linear interpolation of the c lattice parameters of GaN and InN.

sitions of hexagonal $\text{In}_x\text{Ga}_{1-x}\text{N}$. Neither InN or In metal was detected by XRD and TEM studies.

The c lattice parameter was measured by HRXRD for several samples and is plotted in Fig. 1 as a function of the In composition as determined by RBS. The experimental values are found to be greater than values assuming a linear interpolation of the composition with the c lattice parameters of unstrained GaN and InN with $c_0^{\text{GaN}} = 5.1871$ and $c_0^{\text{InN}} = 5.705$ (the dashed line in Fig. 1). The value for x determined from the c lattice parameter would, consequently, result in a composition that is much higher than the actual composition. These values are tabulated in Table I as x , (0002). The in-plane a lattice parameter measured for films with $x = 0.055$, 0.072 , and 0.10 was found to correspond to the a lattice parameter of the underlying GaN, indicating that the films are pseudomorphic and in biaxial compression.

The composition x could be determined from measurements of the lattice parameter from the following expression that includes strain:¹¹

$$\epsilon_c / \epsilon_a = [(c - c_0) / c_0] / [(a - a_0) / a_0] = -2\nu / (1 - \nu), \quad (1)$$

where ϵ_c and ϵ_a is the strain along the c and a axis, respectively, ν is Poisson's ratio, and a_0 and c_0 are the relaxed values of the lattice parameters, which depend on x . The value of ν was found from XRD measurements of GaN to be equal to 0.18.¹² If we assume that the relaxed values are linear in x , i.e.,

$$c_0 = c_0^{\text{GaN}}(1 - x) + c_0^{\text{InN}}(x)$$

and

$$a_0 = a_0^{\text{GaN}}(1 - x) + a_0^{\text{InN}}(x), \quad (2)$$

then the calculated values agree with the composition obtained from RBS (listed in Table I as x , pseudo). This indicates that if biaxial strain is included, the lattice parameter has a linear dependence on composition alloys for $x < 0.10$, and therefore, can be described by Vegard's law.

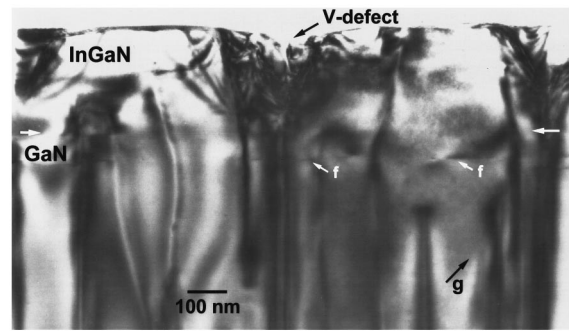


FIG. 2. XTEM micrograph taken near the $[11\bar{2}0]$ zone axis with $g = 1\bar{1}02$ showing no misfit dislocations or additional threading dislocations at an $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}/\text{GaN}$ interface. V grooves observed at the surface of the film are associated with threading dislocations.

For films with $x = 0.114$, the width of the XRD diffraction peaks were too broad to accurately determine the a lattice parameter. However, by using the above conclusion that Vegard's law is valid, the composition was calculated from the measured value of the c lattice parameter and also found to be close to the composition obtained from RBS (x , pseudo in Table I). This indicates that the film is also pseudomorphic with the underlying GaN. The strain was calculated for all the samples by comparing the lattice constants obtained by XRD and the relaxed values from Vegard's law. Table I lists the values of strain and estimated values for the critical thickness from the equation of Matthews and Blakeslee discussed in Ref. 13. It is surprising that these films remain pseudomorphic, given the large amount of strain and the thickness of the films. This suggests the difficulty in the formation of dislocations in $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys at these growth temperatures.

TEM investigations were made to study the dislocation structure at the $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ interface. Figure 2 shows an XTEM micrograph of the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}/\text{GaN}$ interface. The image is taken near the $[11\bar{2}0]$ zone axis with $g = 1\bar{1}02$ in order to reveal the majority of the dislocations that contain both edge and screw components and to enhance the contrast between the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}$ and GaN layer. The horizontal arrows in Fig. 3 mark the position of the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}/\text{GaN}$

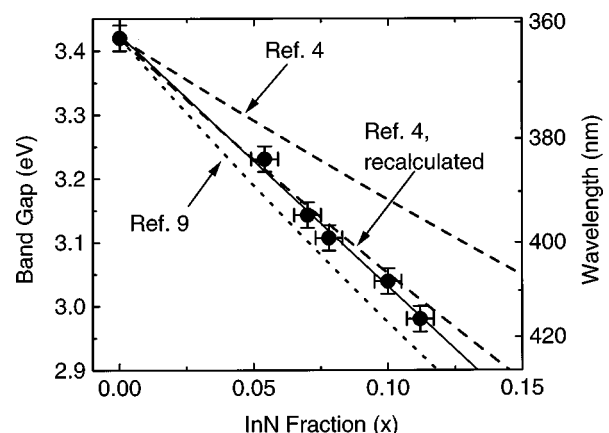


FIG. 3. Measured band gaps of $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys versus InN fraction. The solid line is a least-squares fit to the data of this work. Results of Takeuchi *et al.* (Ref. 9) (dotted line) and Nakamura (Ref. 4) (dashed line) are shown for comparison. We also show a reanalysis of Ref. 4 assuming pseudomorphic strain.

interface. This position was also confirmed with EDX analysis by the absence of the In x-ray line in the GaN layer. The threading dislocations from the underlying GaN are found to propagate through the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}$ layer. No misfit dislocations are detected at the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}/\text{GaN}$ interface. However, within 100 nm of the interface, stacking fault defects (f) are observed in the GaN layer. This was also found in samples of lower composition and in a sample that showed phase separation. A possible origin of the faulted structure is that the large compressive strain in the $\text{In}_x\text{Ga}_{1-x}\text{N}$ layer induces shear in the less compliant GaN layer.

Diffraction contrast (bright/dark regions) along the growth direction can also be observed in the image, which suggests a cell structure that continues from the GaN layer into the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}$ layer. The regions are separated by parallel sets of threading dislocations that are 100–500 nm apart. The surface of the $\text{In}_{0.114}\text{Ga}_{0.886}\text{N}$ layer shows V-groove defects along the $\{10\bar{1}1\}$ planes that are associated with threading dislocations and the cell structure.

The band gaps of the strained $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers were determined by optical transmission spectroscopy and compared to the In composition. Figure 3 shows that our measured values are up to 0.05 eV higher than those previously obtained by Takeuchi *et al.*, who measured photoluminescence (PL) peaks of strained, 40 nm thick $\text{In}_x\text{Ga}_{1-x}\text{N}$ films.⁹ This difference is to be expected, since the PL peak energies of thick $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers are typically lower than the band-gap energies.³ However, our results and those of Takeuchi *et al.* are both in disagreement with previous measurements on thin $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers in MQW structures.^{3,4} In these measurements, only the c lattice parameter was used to determine the composition of the film and therefore resulted in an overestimate of x .^{3,4} We recalculated values for the In composition using $\nu=0.18$ and the assumption of pseudomorphic strain, for the band-gap measurements in Ref. 4. As shown in Fig. 3, the recalculated band gaps are in good agreement with our work and Takeuchi *et al.*, which suggests that the previous study⁴ was performed on pseudomorphically strained $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers. Therefore, this results in a bowing parameter greater than the 1.0 eV that was

originally determined from this study.⁴ Recently, the bowing parameter obtained from our measurements was found to be composition dependent and as high as 3.8 eV for $x=0.1$.¹⁴

In conclusion, we have demonstrated that 225 nm thick $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers grown on GaN are pseudomorphic up to $x=0.114$. This is well beyond the equilibrium critical thickness determined by the lattice misfit. However, TEM showed no misfit dislocations at the $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ interface, which suggests a high activation energy for the formation of dislocations. The band gap of strained $\text{In}_x\text{Ga}_{1-x}\text{N}$ for $x \leq 0.12$ was measured, resulting in a bowing parameter for $\text{In}_x\text{Ga}_{1-x}\text{N}$ that is significantly higher than previously reported.^{3,4,15} Vegard's law was found to be valid for these alloys if the biaxial compression is included.

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