Electron Microscope Study of Strain in InGaN Quantum Wells in GaN Nanowires*

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ABSTRACT

Strains in GaN nanowires with InGaN quantum wells (QW) were measured from transmission electron microscope (TEM) images. The nanowires, all from a single growth run, are single crystals of the wurtzite structure that grow along the <0001> direction, and are approximately 1000 nm long and 60 nm to 130 nm wide with hexagonal cross-sections. The In concentration in the QWs ranges from 12 to 15 at %, as determined by energy dispersive spectroscopy in both the transmission and scanning electron microscopes. Fourier transform (FT) analyses of <0002> and $<1\bar{1}00>$ lattice images of the QW region show a 4 to 10 % increase of the *c*-axis lattice spacing, across the full specimen width, and essentially no change in the *a*-axis value. The magnitude of the changes in the *c*-axis lattice spacing far exceeds values that would be expected by using a linear Vegard's law for GaN – InN with the measured In concentration. Therefore the increases are considered to represent tensile strains in the <0001> direction. Visual representations of the location and extent of the strained regions were produced by constructing inverse FT (IFT) images from selected regions in the FT covering the range of *c*-axis lattice parameters in and near the QW. The present strain values for InGaN QW in nanowires are larger than any found in the literature to date for other forms of $In_xGa_{1-x}N$ (QW)/GaN.

INTRODUCTION

The interesting and technologically promising optoelectronic properties of InGaN quantum wells on GaN have been amply documented [1-3]. The role of strain in controlling the optoelectronic behavior of InGaN-GaN structures has also been well documented [4,5]. Here we report measurements of strain in and around InGaN quantum wells in GaN nanowires made by Bertness *et al.* [6]. This geometry is of interest both for its technological possibilities and for the insight it provides into the behavior of the InGaN-GaN material system.

We describe and apply a novel computer-interactive approach to measuring and imaging local strains using high resolution TEM imaging, based on a Fourier transform (FT) technique. A number of FT methods for measuring strain by use of high resolution lattice imaging have been reported [7-10]. We suggest that our approach is much more physically intuitive and easy to use than the previously reported methods.

PROCEDURES

Experimental

$In_xGa_{1-x}N$ (QW)/GaN nanowires were grown on a (111) Si substrate with molecular beam epitaxy (MBE), with elemental In and Ga and an RF-plasma N_2 source [6]. The substrate temperature was 820 ± 10 °C for the GaN nanowire and 530 ± 20 °C for the $In_xGa_{1-x}N$ quantum wells (QW). The crystallographic structure of the nanowires is wurtzite, with a = 0.3189 nm and c = 0.51856 nm as determined by x-ray diffraction [11]. All of the nanowires investigated were from a single growth run. Approximate local chemical compositions of the QW were obtained by use of energy dispersive spectroscopy (EDS) instruments in both the TEM and the scanning electron microscope (SEM); the In concentration in the QW was 14 ± 2 at %.

The growth axis of the nanowires was normal to the (111) surface of the Si substrate [6]. Transmission electron microscopy samples were prepared by dragging a C-mesh TEM specimen support along the surface. The nanowires had hexagonal cross sections with projected widths of 60 nm to 130 nm. The InGaN quantum well was typically about 80 nm from the end of the nanowire and about 10 nm thick. A TEM image of a typical nanowire including a QW is shown in figure 1.

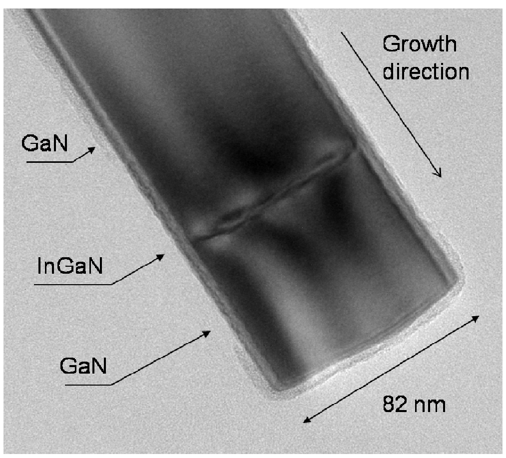

Figure 1. TEM image of the end of a typical GaN nanowire showing the InGaN quantum well near one end. This image is typical of the structure that was imaged at high magnification in the TEM for strain analysis.

TEM imaging was done in two microscopes having very different electron-optical properties [12]. One was a JEOL 2000 FX with coefficient of spherical aberration C_s = 2.3 mm at 200 kV; samples were oriented by use of a single-tilt sample holder. The point resolution at the Scherzer defocus was 0.24 nm. The other microscope was a FEI Titan 80-300 with a C_s-corrected objective; a double-tilt holder was used to orient the specimens. The Scherzer point resolution of the microscope was approximately 0.1 nm at 300 kV.

The growth axis of the nanowires was the c-axis, <0001>. With the single-tilt holder most nanowires could be tilted to an orientation with diffraction vector g = ±0002. In some nanowires, an orientation with g = $\pm 1\bar{1}00$ could also be achieved, but we were not able to obtain the $[11\bar{2}0]$ zone axis orientation. Consequently only lattice fringe images could be obtained. With the double-tilt holder, available in the Titan, we were able to orient nanowires along the $[11\bar{2}0]$ zone axis and obtain lattice structure, or dot, images.

The images were recorded in both microscopes by use of charge-coupled device cameras located below the viewing chamber. In the JEOL TEM the images were recorded at a resolution of approximately 1k x 1k pixels, and in the Titan, 2k x 2k pixels. The original magnification for most images was around 500,000, and exposures of 2 s were typical.

Manual-interactive analysis of TEM images

Fourier transforms (FT) of the high resolution TEM images were obtained by use of either Digital Micrograph, a commercial program, or ImageJ, the PC version of the freeware program NIH Image [12]. FT images from unstrained GaN and graphite were used to calibrate the size of the spot corresponding to the unstrained or reference lattice spacing in the FT image, as well as the spatial frequency associated with the centroid location. The average diameter of the FT spot in a number of such reference images from both microscopes was 4 pixels, so this value was taken as the nominal FT spot size for an unstrained lattice. A three-beam, g = ± 0002, lattice image from an InGaN QW and the associated FT image are shown in figure 2. Careful examination of the FT shows that the (0002) spot is elongated in the axial direction of the nanowire. The spot extends 12 to 15 pixels, implying the presence in the real-space image of a distribution of (0002) lattice spacings, rather than a single uniform lattice parameter.

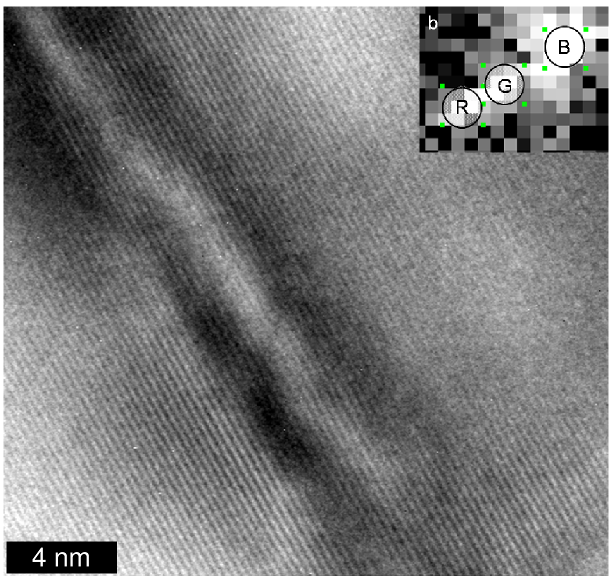

Figure 2. (a) High magnification lattice fringe image, enhanced by Fourier filtering, of an InGaN QW, obtained with three-beam imaging conditions in the JEOL 2000FX. The dark regions are thought to be caused by a variable strain field at the edges of the QW due to non-uniform In concentration. (b, inset) The (0002) spot in the FT of the TEM image, inset, is significantly smeared out from the (0002) center, extending about 12 pixels in (0002) direction. The encircled regions were masked and used to define IFTs that are labeled RGB to follow the color coding used for creating the RGB image shown below in figure 3d.

We used the following method to determine that the lattice parameter differed from its average value in well-defined regions, and thus, to locate these strained regions. At each location of interest in the FT image, a Gaussian mask, 4 pixels in diameter, was placed manually, and the inverse FT (IFT) was performed on the masked region. Each IFT image obtained by this procedure displays the region in the real space image that has a lattice parameter corresponding to the centroid of the mask. For example, figure 2b shows masks at three locations, labeled B, G, and R, in the FT of the image in figure 2a. The associated IFT from each individual mask is shown in figures 3a-3c, respectively. An additive combination of the three IFT images, with the IFTs of the different masks labeled by color, is shown in figure 3d. Comparison of figure 3d with figure 2a shows that the region of largest lattice parameter (red-colored IFT) matches the location of the QW in the original real-space image. This result is consistent with the expectation of localized strain in and near the QW. The location of the centroid of each mask can be determined with sub-pixel precision. In the case of the masks at B, G, and R in figure 3b, the associated (0002) lattice spacings are 0.259 nm, 0.273 nm, and 0.288 nm, respectively.

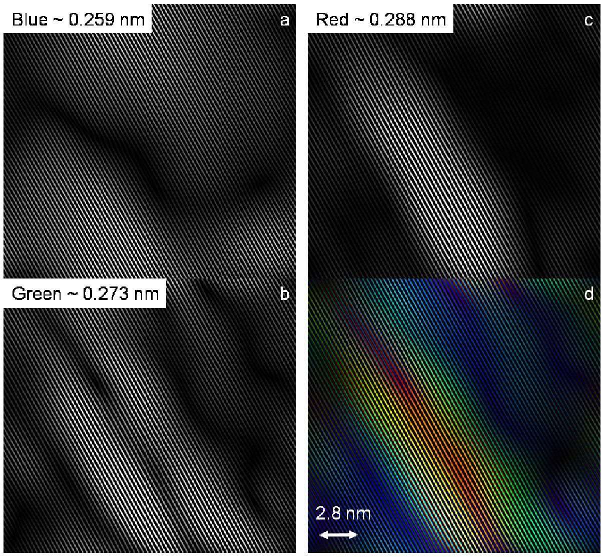

Figure 3. (a) IFT of the spot labeled B in Fig. 2b. This area represents the GaN component of the nanowire. (b) IFT of the spot labeled G in Fig. 2b. This area represents an area with a lattice spacing larger than GaN, and is thought to have a lower In concentration relative to the R spot. (c) IFT of the spot labeled R in figure 2b. This area has the largest lattice spacing. The respective values of the (0002) lattice spacings are indicated in figures 3a, 3b, and 3c. (d) Additive combination of the IFTs in figures 3a, 3b and 3c, with sources indicated by color (in the digital version of this paper). The variation of the (0002) lattice spacings in the QW is thought to be due to non-uniform In concentration.

High resolution TEM lattice structure, or dot, images were taken of GaN nanowires and InGaN QW by use of the C_s-corrected Titan at 300 kV. The image resolution was better than 0.08 nm in these images, as determined by analysis of the FTs. Figure 4a is from an unstrained region of the GaN nanowire and clearly shows the projected atomic columns of Ga in the wurtzite structure. Figure 4b is from the region containing the InGaN QW. The (0002) spots from the FT of both images are included for comparison and show clearly that the (0002) spot in the InGaN image is smeared out compared to the corresponding spot in the FT of the image of GaN only. Close inspection shows the presence of stacking faults and dislocations in the image of the QW, figure 4b, but they should not be the source of the observed smearing. The appearance of the FT and the associated lattice parameter results obtained from images taken by use of the two different TEMs were very similar.

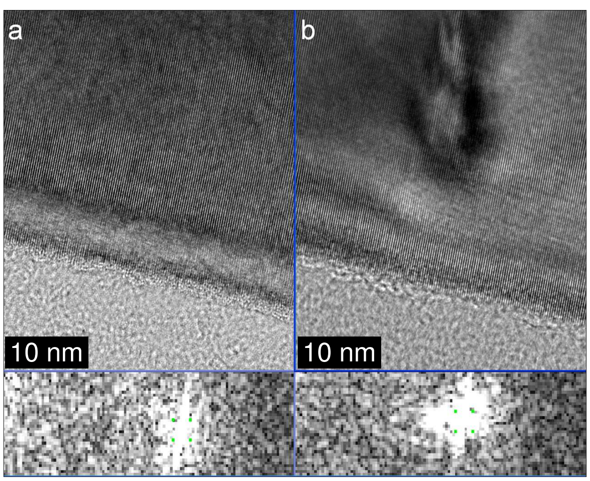

Figure 4. (a) HRTEM lattice structure image (raw image) of an unstrained length of the GaN nanowire and, beneath, the (0002) spot in its FT. (b) HRTEM lattice structure image of a region around an InGaN QW and its (0002) FT spot. Note also the smearing of the (0002) spot in the FT from the QW relative to that from the GaN. The spot in the unstrained FT is about 5 pixels in diameter, while the spot in the QW is about 12 pixels in diameter, or more than two times larger. Analysis shows a maximum strain of 0.053 for this QW.

For each strain measurement, the lattice spacing along the nanowires axis associated with the central region of the QW was tabulated as d_{0002}. For clarity of description, we chose the lattice spacing of the GaN nanowire away from the quantum well as the unstrained reference value for all measurements. This choice is convenient because an accurate unstrained reference value of the lattice parameter throughout the InGaN QW is not known.

Thus, the strain in the <0002> direction is defined as

$$\epsilon_{0002} = \frac{(d_{0002} - d_{GaN})}{d_{GaN}} \quad , \tag{1}$$

where d_{GaN} is the (0002) lattice spacing of GaN, 0.25925 nm [11]. Repeated measurements on single images and measurements on different images of the same specimen indicated that the uncertainty of ϵ_{0002} is about ± 0.01.

RESULTS and DISCUSSION

The maximum strain in the <0002> direction in the InGaN QW shown in figure 2 is 0.11, as determined from the analysis described above and illustrated in figure 3. Results for six different nanowires, along with nanowire diameter, are listed in Table I. Similar analyses of smeared FT spots from images taken with the g = ± $1\bar{1}00$ three-beam conditions show that the strain in directions normal to <0002> is less than 0.005. This is consistent with the expected constraint imposed by the GaN lattice in directions normal to the <0002> growth direction.

Table I. Maximum measured values of axial strain in the InGaN QW (referred to unstrained GaN) and estimates of the strain based on three different physical assumptions.

	Maximum measured values of axial strain from FFT of QW						Vegard's law [13]	Pseudo-morphic [13]	Full volume change, axial strain only
Diameter, nm	60	87	88	102	109	110			
Strain	0.06	0.07	0.07	0.08	0.09	0.13	0.029	0.047	0.093

The present strain values for InGaN in GaN nanowires are larger than any found in the literature for other forms of $In_xGa_{1-x}N$ (QW)/GaN. Görgens *et al.* [13] show that the behavior calculated by a simple application of Vegard's law, that is, simple interpolation between the *c*-axis lattice parameters of unstrained GaN and InN, is quite different from that calculated for the pseudomorphic case, in which the elastic constraint provided by the GaN regions adjacent to the QW has a significant effect. The composition of a few QWs, obtained using EDS with a beam having a 10 nm radius, corresponds to 28±4 % InN in InGaN. This value was used to calculate the strain predicted for the two cases described by Görgens *et al.* [13], using a c-axis lattice spacing of 0.5718 nm [14]; the calculated strain values are listed in Table I.

If one assumes volumetric expansion of the QW according to Vegard's law for unconstrained crystals, and if at the same time its diameter is assumed to be constrained to that of the GaN, then the full volumetric expansion must occur along the nanowires axis. With these assumptions and our In concentration, the axial strain would be 0.093. This purely *ad hoc* model seems to provide an approximate description of the measured strain values.

An explanation for the tendency toward higher strain values for larger diameter nanowires could be pursued by considering the effect of the surface-to-volume ratio or possible non-uniformity in the In content. It has been reported [15] that planar InGaN QW with different thicknesses seemed to have different strains, but the experiments were much different from those of the present study. More measurements on different specimens and more detailed analysis are needed for understanding of the behavior of InGaN QW in GaN nanowires.

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