M R S Internet Journal of Nitride Semiconductor Research

Volume 1, Article 6

The Morphology and Cathodoluminescence of GaN Thin Films

Carol Trager-Cowan , P. G. Middleton, K. P. O'Donnell
Department of Physics and Applied Physics, University of Strathclyde

This article was received on May 31, 1996 and accepted on September 5, 1996.

Abstract

In this paper we compare gallium nitride (GaN) films grown by molecular beam epitaxy on sapphire (Al_2O_3), gallium arsenide (GaAs (111)B) and lithium gallate ($LiGaO_2$) substrates. Atomic force microscopy, scanning electron microscopy, cathodoluminescence imaging and cathodoluminescence spectroscopy are used to characterise the films. From growth runs carried out to date, GaN films on GaAs substrates exhibit the best surface uniformity and the cleanest luminescence.

1. Introduction

The commercialisation of light emitting diodes based upon InGaN [1] has resulted in an explosion of interest in wide bandgap III-V materials. The ability to produce light emitting diodes spanning the UV to orange spectral region makes these materials highly attractive for the development of full colour displays. Recent announcements by Nagoya Institute of Technology [2], of stimulated emission at 380 nm from a InGaN/AlGaN diode at room temperature, and by Nichia Chemicals, of a laser diode operating at 410 nm [3], has generated further interest in developing coherent light emitters for high density optical data storage, reprographics, underwater communications and so on.

In spite of recent successes, much fundamental physics remains to be done on these materials. Working devices exhibit huge defect densities [4]. Unidentified deep centres contribute a strong green/yellow band in competition with band edge emission. We use atomic force microscopy, scanning electron microscopy, cathodoluminescence imaging and cathodoluminescence spectroscopy, to study both the morphological and optical properties of GaN films and to investigate the correlation between them. In this paper we compare results obtained from MBE grown GaN on three different substrates: (0001) orientated sapphire (Al_2O_3), gallium arsenide (GaAs (111)B) and the novel lattice-matched oxide substrate lithium gallate ($LiGaO_2$).

2. Sample Growth

All the films discussed in this paper were grown by molecular beam epitaxy using a solid source for gallium and an RF plasma source for atomic nitrogen [5]. Prior to growth, the sapphire substrates were degreased using standard solvents. They were then thermally heated at ~800^{o}C for 20-30mins and finally lowered to the growth temperature of 750^{o}C. The epi-ready GaAs(111)B substrates were thermally cleaned at ~600^{o}C to remove the surface oxide and were nitrided for a few minutes as the temperature was raised to the growth temperature of 700^{o}C. As for the $LiGaO_2$ substrates, these were etched in a solution of phosphoric and suphuric acids HPO_4:H_2SO_4 (1:1) at 80^{o}C for about 10-15 mins before introducing them into the MBE system. The gallate was then nitrided as the substrate temperature was increased to 630^{o}C for 30mins before initiating the growth. No buffer layers were deposited prior to GaN epilayer growth.

3. Experimental Techniques

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press

3.1. Atomic Force Microscopy

A Burleigh Personal atomic force microscope operating in ambient conditions provides high-resolution physical imaging of samples within a frame up to 70 μm square. In this paper we present atomic force microscopy (AFM) images, either 1.4 μm or 7.0 μm square, acquired with a resolution of 256 x 256 pixels. Scan times of approximately 10 minutes are necessary to obtain high quality images.

3.2. Scanning Electron Microscopy and Cathodoluminescence Imaging

A scanning electron (SE) microscope has been modified to enable light, i.e., cathodoluminescence (CL), to be detected as the electron beam is scanned over the surface of a sample. The resultant CL micrograph can be compared with a SE micrograph obtained from the same area of the sample, allowing a direct comparison of the optical morphology of a sample with its physical morphology. By inserting appropriate absorption filters or a monochromator into the luminescence exit beam, CL images that correspond to selected wavelengths of emission are readily obtained. SE and CL micrographs are acquired by digitisation of the images by a frame grabbing card installed in a personal computer.

3.3. Cathodoluminescence Spectroscopy

A home-built cathodoluminescence spectrometer provides an electron beam with energies up to 30 keV at current densities to ≈ 20 A/cm^2 (but limited in the work to be described here to less than 20 mA/cm^2). The electron beam excitation spot may be focussed to a region of diameter 200 μm, or defocussed to illuminate a whole sample, up to a diameter of about 5 mm. Spectral acquisition in the range of photon energies from 2.0 to 3.5 eV utilises either a R928 Hamamatsu photomultiplier tube or an Oriel InstaSpec^{TM} cooled 2-dimensional CCD array mounted at the output focal plane of a Chromex 0.5M monochromator. Use of the CCD array brings the following advantages: (1) Entire spectra are obtained very quickly. Acquisition times in the present work range from 0.5 to 40 seconds. (2) Some limited spatial resolution is achieved within the excited region of the sample. The CCD comprises 1024 by 256 pixels, with each pixel measuring 27 μm square. The luminescence from the sample is dispersed in the horizontal plane and "imaged" in the vertical plane. A single spectrum may be acquired by integrating the signals over the rows of the CCD. Alternatively *n*-spectra ($1 \le n \le 256$) may be acquired by dividing the array via software into vertical tracks. Spectra presented in this paper were acquired by dividing the array into 4 (1024 x 64 pixels) tracks or 8 (1024 x 32 pixels) tracks. With the luminescence collection optics providing a magnification equal to 1.4, and the entrance slit width of the monochromator set to 40 μm, spectra were obtained from slices of sample ≈ 30 μm wide by ≈ 1.2 mm high and ≈ 30 μm wide by 0.6 mm high respectively.

4. Results

4.1. Atomic Force Micrographs

The atomic force micrographs presented in Figure 1 highlight the considerable differences that exist in surface morphology between GaN films grown by MBE on different substrates. The GaN/Al_2O_3 film comprises distinct hexagonal crystallites, often showing central depressions on the top face, that stand clear of an ill-defined background. All microcrystals are of roughly the same size, 1 to 3 μm across a hexagonal face and about 1 μm high. The GaN/$LiGaO_2$ film also appears to comprise a matrix of crystallites but the largest crystallites are only about 1 μm in diameter and are less than 300 nm high. These are superimposed on a background of even smaller crystallites (around 250 nm diameter and a few 10's nm high). In sharp contrast, the GaN/GaAs (111)B film shows a uniform, quasi-smooth surface: note the reduced scan size of 1.4 μm square, and the reduced vertical scale in Figure 5c. Some ill-defined crystallites of diameter around a quarter of a micron and height less than 50 nm can be discerned in the GaN/GaAs (111)B image.

4.2. Scanning electron microscopy and cathodoluminescence imaging

Figure 2 shows comparative scanning electron microscopy (SEM) and panchromatic CL images from a GaN film grown on Al_2O_3. The CL image shows a strong contrast between brightly luminescing crystallites and a uniformly dark background.

In the geometry used for CL imaging, with the sample surface normal to the exciting electron beam, no discernible structure was observed in SEM and CL images of GaN films grown on the GaAs (111)B and $LiGaO_2$ substrates. In

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press

contrast, AFM images clearly show vertical structure in these films. This comparison emphasises the greater vertical as well as spatial resolution obtainable using AFM compared to standard SEM.

4.3. Cathodoluminescence Spectroscopy

4.3.1. GaN/Al_2O_3

Figure 3 shows low temperature spectra acquired at 5 keV from several different regions of a GaN film grown on a Al_2O_3 substrate using a PMT mounted at the output of the monochromator. There are striking variations from point to point on the sample. The ratio of UV band edge to deep level yellow emission depends critically on the beamspot position, as does the appearance of blue emission in the region near 3.0 eV.

4.3.2. $GaN/LiGaO_2$

Figure 4 shows low temperature spectra from a 0.5 μm thick $GaN/LiGaO_2$ film acquired using a range of electron beam energies, at different positions within the excitation spot. These spectra were acquired with the CCD array divided into 8 tracks. Figure 4 shows spectra acquired in tracks 3, 5, 6 and 7. From this figure we can deduce that (1) as demonstrated for the GaN/Al_2O_3 film, the $GaN/LiGaO_2$ emission properties are non-uniform and (2) with increasing electron beam energy (i.e., probing deeper into the film) the ratio of UV band edge emission to yellow deep level emission decreases. The maximum range (Bethe range) of a 2keV electron beam in GaN was deduced from Monte-Carlo simulations of the electron trajectories [6] to be ≈ 40 nm, the Bethe range of a 7keV electron beam was calculated to be ≈ 300 nm.

4.3.3. GaN/GaAs (111)B

Figure 5a shows low temperature spectra from a 0.8 μm GaN/GaAs (111)B film. These spectra were acquired with the array divided into 4 tracks. Figure 5b shows low temperature spectra acquired using different electron beam energies to probe different depths of the film. The Bethe range of a 3 keV beam in GaN was calculated to be ≈ 80 nm, the Bethe range of a 15keV beam was calculated to be ≈ 1.2 μm. From this figure we can deduce: (1) This sample exhibits the most uniform luminescence of the three samples studied; (2) The ratio of the UV band edge emission to yellow deep level emission varies only weakly with depth.

The inset in figure 5b shows that the UV band edge emission shifts to higher energy as the electron beam energy is reduced. This result indicates variation of strain in the layer with depth. As the lattice mismatch between GaN and GaAs (111)B is very large (25% at room temperature), it is reasonable to assume that the strain due to lattice mismatch relaxes during growth of a 0.8 μm layer. A variation in strain in the layer may result from the difference in thermal expansion coefficients between GaN (5.6×10^{-6}/K) and GaAs (5.87×10^{-6} /K). As the thermal expansion coefficient for GaAs is greater than that for GaN, a layer cooled from growth temperature is subjected to compressive strain. The blue shift of the band edge emission as we approach the surface is consistent with a relaxation of this compressive strain. Piezoelectric fields may enhance the bandedge shift.

5. Summary

Comparative morphological and spectroscopic studies of GaN films grown on Al_2O_3, GaAs (111)B and $LiGaO_2$ substrates clearly show that the GaN/GaAs (111)B film exhibit the best surface uniformity and cleanest luminescence. AFM, SEM, CL imaging and CL spectroscopy prove to be very useful techniques for characterising the heterogeneous material available at the present state of the art.

Acknowledgments

The $LiGaO_2$ substrates were grown by John Nicholls of the Optical Materials Research Centre at Strathclyde University headed by Professor Brian Henderson. Work at Nottingham University on the GaN/Al_2O_3 films was funded by Sharp Laboratories of Europe (SLE) Ltd.. The University of Strathclyde is grateful to SLE for the loan of these samples. We thank Susanne Craig and Billy McCallum for producing the AFM micrograph and CL spectra, respectively from the GaN/Al_2O_3 films. We thank Graham Watt for his work interfacing the SEM. We thank David Clark, Ged Drinkwater and Jim Barrie for laboratory assistance. CL spectroscopy at Strathclyde is funded by EPSRC/MOD grant GR/H04404 and EPSRC

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press

grant GR/K16968.

References

[1] Shuji Nakamura, Takashi Mukai, Masayuki Senoh , *Appl. Phys. Lett.* ***64***, 1687-1689 (1994).

[2] I Akasaki, H Amano, S Sota , H Sakai, T Tanaka, M Koike, *Jpn. J. Appl. Phys.* ***34***, 1517-1519 (1995).

[3] S Nakamura, M Senoh, S Nagahama, N Iwasa, T Yamada, T Matsushita, H Kiyoku, Y Sugimoto, *Jpn. J. Appl. Phys.* ***35***, L74-L76 (1996).

[4] S. D. Lester , F. A. Ponce , M. G. Craford, D. A. Steigerwald , *Appl. Phys. Lett.* ***66***, 1249-1251 (1995).

[5] T. S. Cheng, L. C. Jenkins, S. E. Hooper, C. T. Foxon , J. W. Orton, D. E. Lacklison , *Appl. Phys. Lett.* ***66***, 1509-1511 (1995).

[6] E Napchan, D B Holt, *Inst. Phys. Conf. Ser.* ***87***, 733-738 (1987).

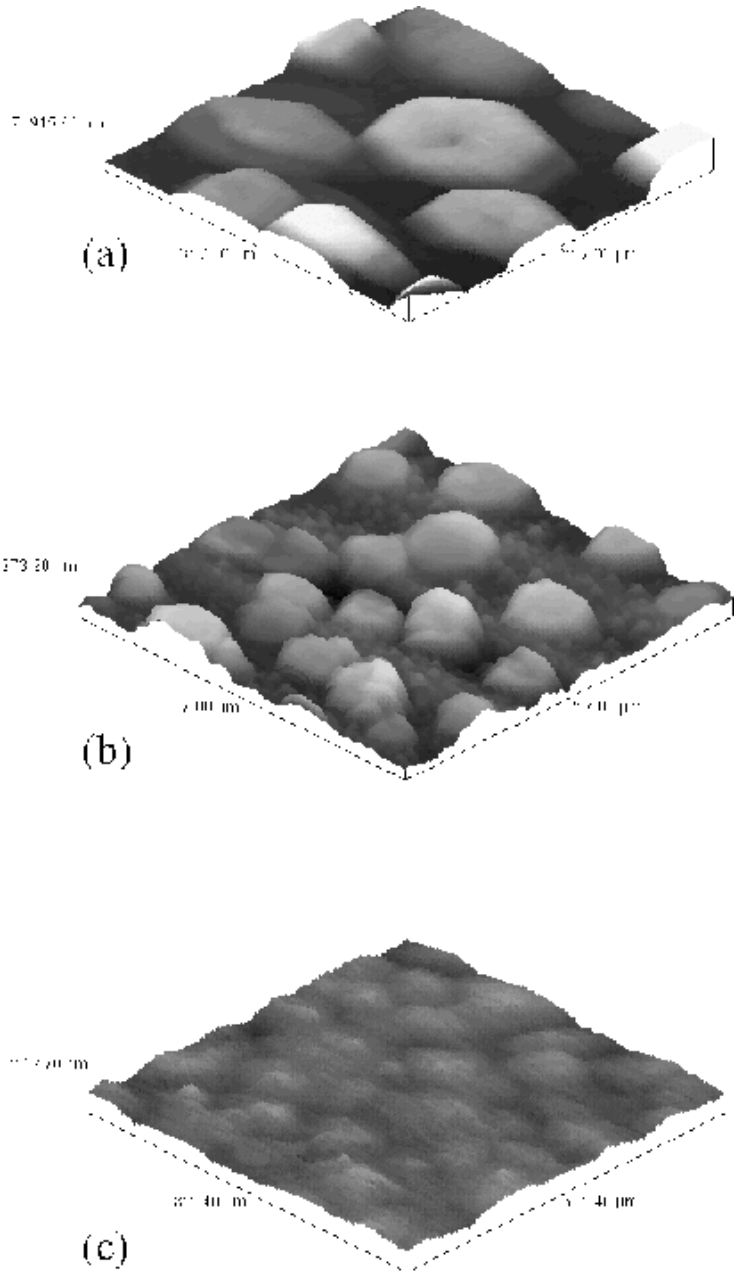

Figure 1. AFM images of (a) GaN/Al_2O_3 (b) GaN/$LiGaO_2$ (c) GaN/GaAs (111)B

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press

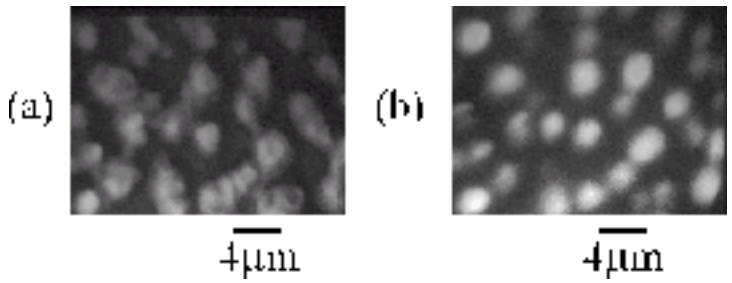

Figure 2. Comparing (a) a SE micrograph with a (b) panchromatic CL image of a GaN/Al_2O_3 film.

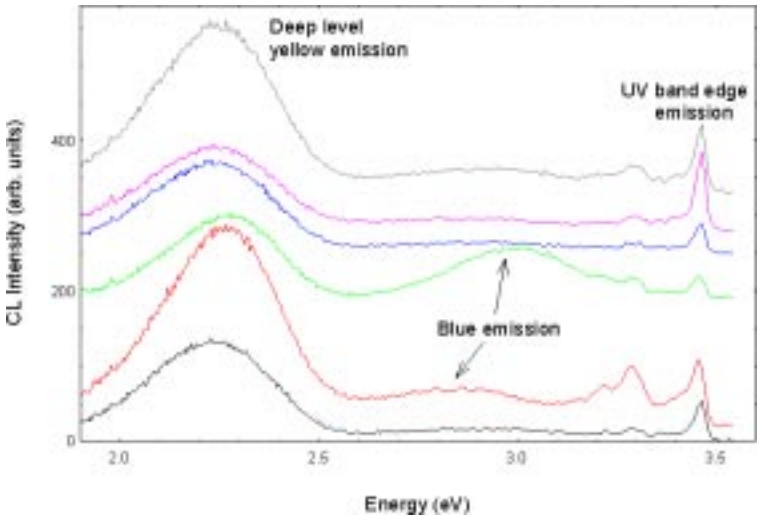

Figure 3. CL spectra obtained from different points on a GaN/Al_2O_3 film.

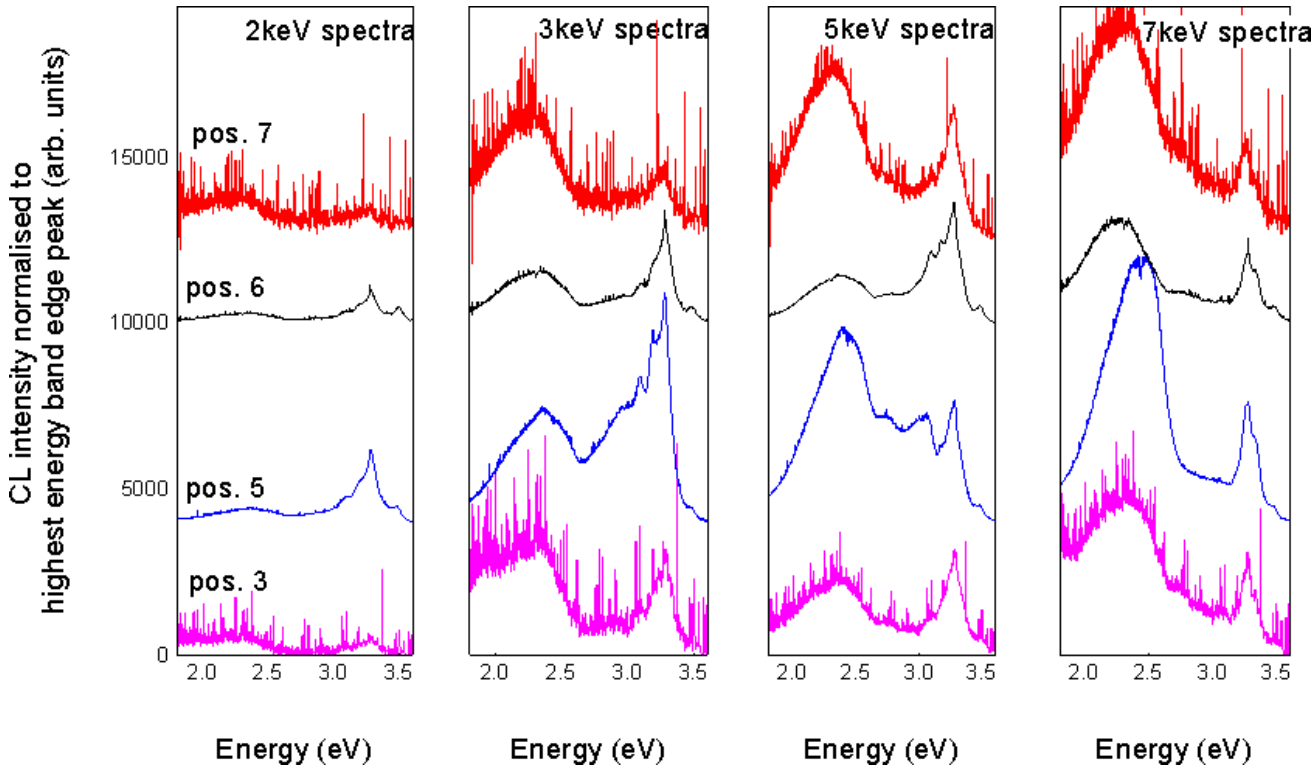

Figure 4. CL spectra acquired at different positions within the excitation spot for a range of electron beam energies for a GaN/$LiGaO_2$ film.

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press

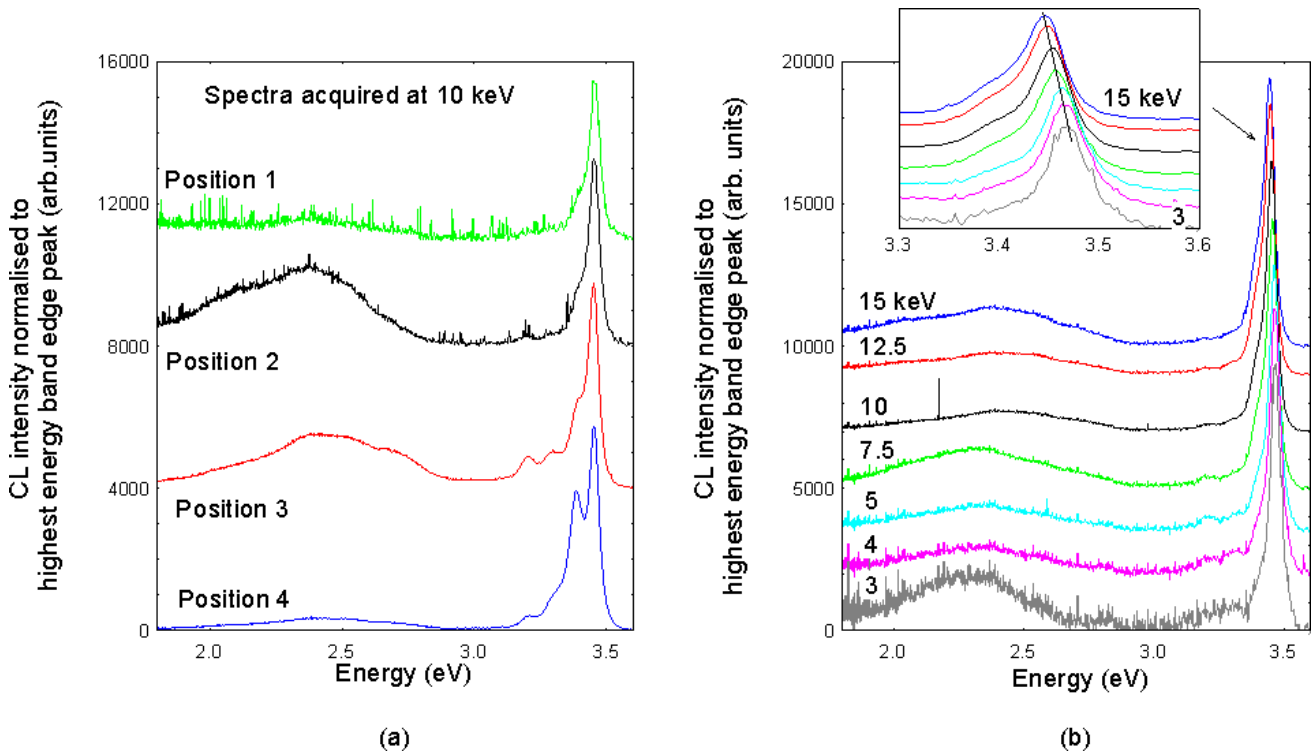

Figure 5. CL spectra acquired at (a) different positions within the excitation spot and (b) for a range of electron beam energies for GaN/GaAs (111)B film.

M R S Internet Journal of Nitride Semiconductor Research

https://doi.org/10.1557/S1092578300001782 Published online by Cambridge University Press