Superlattices and Microstructures 45 (2009) 151–155

Contents lists available at ScienceDirect

Superlattices and Microstructures

journal homepage: www.elsevier.com/locate/superlattices

Correlating composition and luminescence in AlInGaN epilayers

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ARTICLE INFO

Article history:
Available online 25 November 2008

Keywords:
AlInGaN
GaN
MBE
Cathodoluminescence
WDX

ABSTRACT

Epilayers of the quaternary alloy $Al_xIn_yGa_{1-x-y}N$ have been grown on GaN/sapphire templates by plasma-assisted molecular beam epitaxy. The emission properties and elemental compositions of these samples were evaluated simultaneously and intercorrelated by combining hyperspectral cathodoluminescence imaging and wavelength-dispersive X-ray mapping. Use was made of inherent variations in growth temperature across a single epilayer to study the resultant effect on the different metal fractions and luminescence emission wavelength. By examining statistical correlations in this data, the interdependence of the fractions of constituent binary compounds, together with the associated changes in emission characteristics, can be clarified without the need to grow a systematic series of samples.

1. Introduction

The quaternary alloy $Al_xIn_yGa_{1-x-y}N$ (hereafter referred to as AlInGaN) holds promise for a number of uses, particularly in applications where it can be lattice-matched to GaN [1,2] with a variable UV band-gap. However, better understanding of the growth mechanism of this material is an essential prerequisite for its technological development, particularly given the conflicting growth conditions required by the constituent binary compounds. Optimisation of growth temperature is crucial since InN requires a relatively low growth temperature, whilst GaN prefers a higher temperature and AlN higher still. Previous work by some of the present authors [3–5] has focused on the analysis of series of samples, each grown under marginally different conditions. In the current work, we exploit the

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doi:10.1016/j.spmi.2008.10.014

temperature-induced composition variation commonly present across a single epilayer, combined with a large number of spatially resolved measurements, in order to investigate such variation statistically. This method has been successfully applied to a number of different epilayers, a single example of which is presented in this work.

2. Sample growth

The AlInGaN samples were grown by plasma-assisted molecular beam epitaxy (PA-MBE) in a Riber Compact 21 reactor, using an Addon r.f. plasma N source and Al, Ga and In Knudsen cells. A 100 nm buffer layer of GaN was initially grown at low temperature (730 °C) on a Lumilog *c*-plane GaN-on-sapphire template. This was followed by ≈300 nm of wurtzite AlInGaN grown at a temperature of 595 °C under intermediate metal-rich conditions to ensure a two dimensional growth and to avoid the formation of metal droplets on the surface [5]. The rotation centre during the growth was in the centre of the sample, which was one-quarter of a 2-in. wafer. Due to edge effects, there was a temperature gradient across the sample during growth with a higher temperature close to the edges.

3. Measurement details

The composition and luminescence spectra of the samples were measured in a combined cathodoluminescence (CL) and wavelength-dispersive X-ray (WDX) spectroscopy system. Based on a Cameca SX100 electron probe microanalyser, this instrument makes use of an optical microscope which is co-axial and confocal with the electron beam to collect light emitted from the sample under electron beam excitation [6]. At each point in the rastered scan, a complete room-temperature CL spectrum is recorded using a CCD spectrograph, resulting in a multidimensional data cube, or hyperspectral image. Elemental data is acquired simultaneously using three WDX spectrometers, allowing composition and emission to be mapped together under identical conditions and avoiding issues of position registration which would otherwise arise from use of non-simultaneous *ex-situ* measurements.

Beam parameters were chosen to yield a suitable signal/noise ratio whilst confining the excitation to the quaternary epilayer. The 6 kV, 20 nA beam was held stationary whilst the sample was scanned in 50 μm steps over an area of 4.05 × 9.45 mm, taking 800 ms per pixel. Ga Lα and Al Kα X-rays were detected with WDX spectrometers using thallium acid phthalate (TAP) diffracting crystals, with a pentaerythritol (PET) crystal used to monitor the In Lα line. Twenty points were measured across the sample using fully quantitative WDX (i.e. using standards-based, ZAF-corrected peak-minus-background analysis), and these were used to calibrate the X-ray count maps in terms of AlN, GaN and InN fractions [7]. Nitrogen content was also measured for these points, and the material was confirmed to be stoichiometric at all measured points. Rutherford backscattering spectrometry measurements carried out on similar samples showed good agreement with the WDX analysis [5].

4. Results

Fig. 1 shows the results of mapping the In, Ga and Al X-ray peak counts after scaling them with the quantitative data. The InN and GaN maps clearly show the effect of a growth temperature gradient across the sample, with increasing InN and decreasing GaN content towards the top right of the maps; this corresponds with a lower temperature in this region, which was near the centre of the growth wafer. Little variation in the AlN content is observed; this is despite the fact that it would be expected to show a similar behaviour to GaN, due to near-unity sticking fractions for both Al and Ga in MBE growth in this temperature region. This behaviour is analysed in depth in Ref. [3]. Fig. 2 shows an alternative representation of the composition data, in the form of a ternary plot showing the relative abundance of the three binary compounds in the alloy, in which each point represents one of the 15,580 pixels in the X-ray maps with the sum of the binary compositions set to 100%. The data are seen to form an elongated cluster almost parallel to the lines of constant AlN. There is, however, a slight tendency for the AlN content to increase with increasing GaN, albeit at a slower rate, as illustrated by the major and minor axes of the ellipse.