Journal of Crystal Growth 378 (2013) 295–298

Contents lists available at SciVerse ScienceDirect

Journal of Crystal Growth

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

Study of the pseudo-(1×1) surface by RHEED and XPS for InGaN/GaN (0001)/Al_2O_3 heterostructures grown by PA-MBE

E. Cruz-Hernández [a,*], M. Ramirez-Lopez [b], M. Pérez-Caro [b], P.G. Mani-Gonzalez [c,d], A. Herrera-Gómez [c], A. Yu Gorbatchev [a], M. López-López [b], V.H. Méndez-García [a]

[a] Center for the Innovation and Application of Science and Technology (CIACyT), Universidad Autónoma de San Luis Potosí, Sierra Leona 550, Lomas 2a Secc, C.P. SLP 78210, México
[b] Physics Department, CINVESTAV-IPN. D.F., México
[c] CINVESTAV- Unidad Querétaro, México
[d] Instituto de Ingeniería y Tecnología, Departamento de Física y Matemáticas, Universidad Autónoma de Ciudad Juárez, Ave. del Charro 450, Cd. Juárez C.P. Chihuahua 32310, México

ARTICLE INFO

Available online 20 January 2013

Keywords:
A1. Diffusion
A1. Reflection high energy electron diffraction
A1. Nanostructures
A3. Molecular beam epitaxy
B1. Nitrides

ABSTRACT

In this letter, we report on the study of the pseudo-(1×1) surface by reflection high-energy electron diffraction in the InGaN grown on GaN/sapphire substrate by plasma-assisted molecular beam epitaxy. At the end of the InGaN layer growth at 500 °C the formation of a (1×1) diffraction pattern along the [11–20] azimuth was observed. By decreasing the temperature to 330 °C distinct sidebands appear on the high wave vector sides of these first-order streaks. The observation of this kind of 1/6-satellite peaks is then related with the formation of an outmost metal bilayer. From atomic force microscopy, nanometer-size surface structures consisting of hexagonal rows are observed. X-ray photoelectron spectroscopy analysis reveals relatively smaller concentration of Ga as compared with In within the surface area than in the bulk of the InGaN layers. We explain the results by means of the surface In segregation and proposed a possible model for the In-bilayer atomic surface distribution.

1. Introduction

For the last years, compound such as GaN and Ga-rich InGaN have been successfully used in a diversity of advanced photonic and optoelectronic devices operating in the blue and ultraviolet range of the electromagnetic spectrum. Since the bandgap of $In_xGa_{1-x}N$ alloys can be varied from 0.7 to 3.4 eV, covering completely the visible spectrum, they are of special interest for potential optoelectronic applications such as light emitting diodes (LEDs) or laser diodes (LDs) [1,2]. Besides, due to its high carrier mobilities, high absorption coefficients or high heat capacity and low sensitivity to ionizing radiation the $In_xGa_{1-x}N$ has high potential to be applied in the solar energy industry [3,4]. Additionally, the small effective mass and high mobility in the high In composition $In_xGa_{1-x}N$ are important for potential high speed device applications [5]. On the other hand, Smith et al. reported that in the Ga-rich surface reconstruction of GaN (0001) is observed by reflection high-energy electron diffraction (RHEED) mainly 1×1 streaks at the growth temperature but, as the surface is cooled down to < 350 °C, distinct sidebands appears on the high wave vector sides of the first-order streak along the [11–20] azimuth [6]. The streak splitting was termed "1×1" or pseudo-(1×1) surface. That kind of surface contains 2–3 additional monolayers of Ga above the Ga-terminated bilayer. Recently, it has been observed standing waves at steps on this pseudo-(1×1) surface by scanning tunneling spectroscopy at room temperature showing that the structure is nearly free-electron-like, thus confirming the metallic state of this surface [7].

In this letter, we describe a similar effect observed in the system InGaN/GaN(0001)/sapphire grown by Plasma-assisted Molecular Beam Epitaxy (PA-MBE), where originally (1×1) RHEED streaks evolve, after cooling down the sample, to a splitting pattern of a pseudo-(1×1)-like surface. By X-ray photoelectron spectroscopy (XPS) results and physical considerations, this splitting is explained as due to In segregation towards the surface.

2. Experimental procedure

The growth was carried out in a Riber C21 MBE system on Al_2O_3 wafers. After the substrate nitridation process performed at 800 °C the substrate temperature was set at 550 °C where a GaN layer was deposited for 10 min. The sample was annealed at 800 °C for 20 min, and then a 20-period AlN/GaN super lattice was

* Corresponding author. Tel.: +52 444 4448 262 300.
E-mail addresses: esteban.cruz@uaslp.mx, echzeroth@gmail.com (E. Cruz-Hernández).

0022-0248/$ - see front matter
http://dx.doi.org/10.1016/j.jcrysgro.2012.12.172

grown at 780 °C followed by a high temperature (750 °C) GaN layer grown by 2 h. Finally, a 4-minutes (40–50 nm) film of InGaN was deposited at 500 °C. Immediately after the InGaN deposition, all fluxes were shut down and then the substrate temperature was lowered down to room temperature. The sample was grown under N-rich conditions and the indium content incorporated in the crystalline lattice was set to x=0.25. In-situ analysis was carried out by reflection high-energy electron diffraction (RHEED). The surface morphology of the samples was studied by atomic force microscopy (AFM) at room temperature in contact mode. High-resolution XPS measurements were performed using a Thermo Fisher-VG instruments system equipped with a monochromatic Al Kα (1486.7 eV) X-ray source and a hemispherical electron analyzer with seven channeltrons. To optimize resolution, the pass energy employed was 10 eV. The base pressure of the analysis chamber was 7×10^{-10} Torr and increased to 3×10^{-10} Torr of data acquisition. The electron take-off angle was controlled by tilting the sample; the angles employed correspond to 90° and 45° measured from the surface. The XPS core-level peaks were deconvoluted into their various components by using an interactive least-squares computer program and fitting to Gaussian curves.

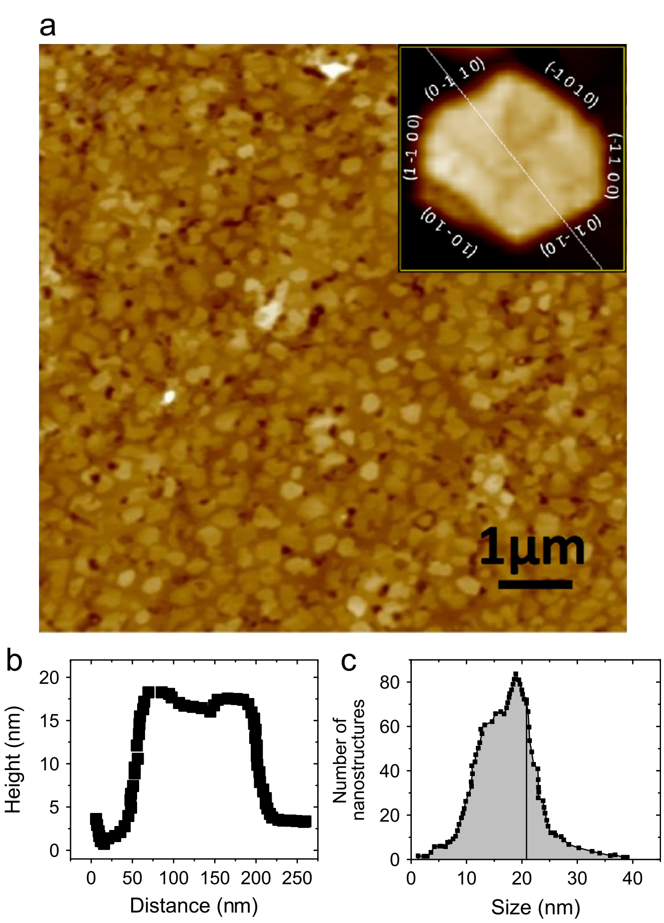

Fig. 1. (a) 8 µm × 8 µm AFM image of the surface morphology observed at the end of the InGaN growth. A dense array of hexagonal nanostructures is obtained. Inset: detail of one of these hexagonal islands bounded by (0−110), (−1010), (−1100), (01−10), (10−10) and (1−100) planes as analyzed by AFM. (b) Height profile taken along the hexagonal structure of the inset in (a). (c) Size distribution of the nanostructures taken from the AFM image.

3. Results and discussion

Under the described growth conditions, atomic force microscopy (AFM) images reveal the formation of hexagonal nanostructures of average transversal length of 120 nm as can be seen in Fig. 1(a). In the inset it is shown a close look of one of these structures and the planes that define the sides of the hexagonal prism. Fig. 1(b) shows a height profile from the AFM image taken along the hexagonal structure, as depicted in the inset. Fig. 1(c) indicates that the average height of the structures is ~20 nm. The spontaneous formation of this kind of superficial morphologies can be explained as driven by the significant strain in the surface layers and due to the relative weakness of the In–N bond compared to the Ga–N bond [8].

From RHEED patterns taken along the [11–20] azimuth, at T_g=750 °C, it is confirmed the 1 × 1 reconstruction at the end of the GaN layer growth as expected for GaN (0001) surface [6]. At the end of the InGaN layer growth the preservation of the 1 × 1 diffraction pattern was observed, which is associated to a stable ternary structure as reported elsewhere [9]. However, by decreasing the temperature to 330 °C distinct sidebands appear on the high wave vector sides of the first-order streaks along the [11–20] azimuth, as shown in Fig. 2(a). An intensity profile taken from RHEED pattern of Fig. 2(a) is shown in Fig. 2(b). In there, the splitting of the original streaks is evident, that is, the initial 1 × 1 surface reconstruction evolves towards a reconstruction with satellite streaks. The spacing between these additional features is located approximately at 1/6 from the zeroth- and first-order streaks. Since the 1/6-satellites were not observed at the InGaN growth temperature (500 °C), these results suggest that In surface segregation is triggered as the sample was cooled down to 330 °C. Indium segregation is a common issue that has been previously reported for this system [8] and has been explained in terms of

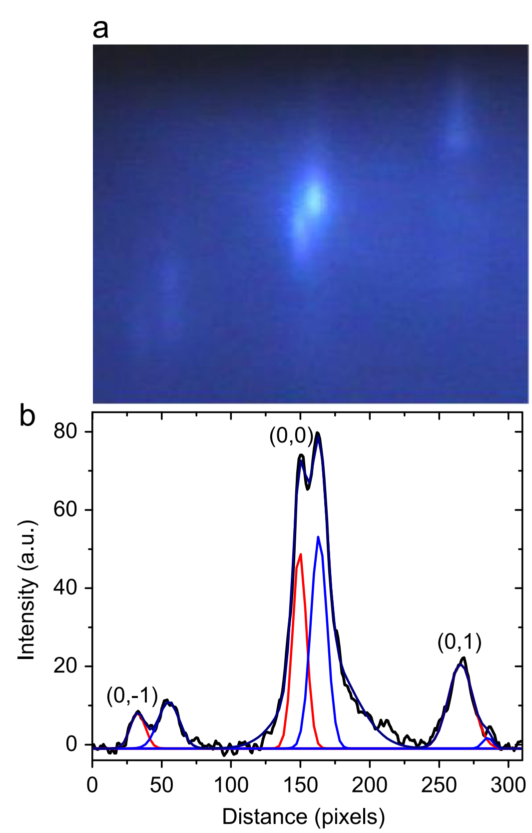

Fig. 2. (a) RHEED pattern observed along the [11–20] direction showing the pseudo-(1 × 1) InGaN/GaN (0001) surface reconstruction. (b) RHEED-intensity profile where the double rods can be better identified.