Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of Crystal Growth 270 (2004) 346–350

JOURNAL OF CRYSTAL GROWTH

www.elsevier.com/locate/jcrysgro

GaN/GaAs(1 0 0) superlattices grown by metalorganic vapor phase epitaxy using dimethylhydrazine precursor

J. Sormunen*, J. Riikonen, M. Sopanen, H. Lipsanen

Optoelectronics Laboratory, Helsinki University of Technology (HUT), P.O. Box 3500, FIN-02015 Espoo, Finland

Received 17 March 2004; accepted 30 June 2004
Communicated by D.W. Shaw

Abstract

Superlattices of cubic gallium nitride (GaN) and gallium arsenide (GaAs) were grown on GaAs(1 0 0) substrates using metalorganic vapor phase epitaxy (MOVPE) with dimethylhydrazine (DMHy) as nitrogen source. Structures grown at low temperatures with varying layer thicknesses were characterized using high resolution X-ray diffraction and atomic force microscopy. Several growth modes of GaAs on GaN were observed: step-edge, layer-by-layer 2D, and 3D island growth. A two-temperature growth process was found to yield good crystal quality and atomically flat surfaces. The results suggest that MOVPE-grown thin GaN layers may be applicable to novel GaAs heterostructure devices.

PACS: 81.15.Gh; 68.55.Jk

Keywords: A1. Nanostructures; A1. X-ray diffraction; A3. Metalorganic vapor phase epitaxy; A3. Superlattices; B1. GaAs; B1. GaN

1. Introduction

In recent years, high-quality gallium nitride (GaN) has been grown on a variety of substrates to yield a range of electronic and optoelectronic applications, including the blue laser diode [1]. The cubic zinc blende phase of GaN on substrates such as GaAs(1 0 0) is considered to be suitable for some heterostructure applications [2,3]. Despite the problems in the growth of GaN resulting from the large lattice mismatch (~20%) between the materials, a few nanometers of GaN on GaAs has been shown to be smooth [4,5]. As the critical thickness of cubic GaN on GaAs is less than one monolayer, these layers are assumed to be relaxed by Lomer dislocations [6]. By growing GaAs on the deposited GaN, a barrier layer with a large valence band offset of approximately 1.8 eV for holes can be achieved [7]. Thus, nanoscale GaN/GaAs heterostructures might be applicable to

---

*Corresponding author. Tel.: +35894515312; fax: +35894513128.

E-mail address: jaakko.sormunen@hut.fi (J. Sormunen).

0022-0248/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.jcrysgro.2004.06.055

components such as the resonant tunneling diode (RTD), realized so far from, e.g., SiGe/Si [8,9] and GaAs/AlAs [10]. To fabricate a device-quality GaN/GaAs structure, the growth temperature of GaN should be fairly low as decomposition of GaAs at 800 °C and above leads to rapid degradation of crystal quality [11]. In this paper, we present a 5-period GaN/GaAs superlattice (SL) grown by metalorganic vapor phase epitaxy (MOVPE). The SL is a test structure used to study the growth of layers that may be applied to typical tunneling components. A two-temperature process is employed to optimize the growth conditions for the nitride and arsenide layers of the SL. We also characterize how the crystalline quality and surface morphology of the structure depend on the GaN layer thickness and growth temperature.

2. Experimental procedure

The samples were fabricated in a horizontal MOVPE reactor at atmospheric pressure using hydrogen as carrier gas. Tertiarybutylarsine (TBAs), trimethylgallium (TMGa), and dimethylhydrazine (DMHy) were used as precursors for arsenic, gallium, and nitrogen, respectively. Due to its low decomposition temperature, DMHy allows growth at temperatures around 550 °C [12]. Prior to the growth of the superlattice semi-insulating GaAs(1 0 0) substrates were annealed at 700 °C to remove the native oxide. Additionally, a 50-nm-thick GaAs buffer layer was deposited at 670°C. The temperatures reported here are thermocouple readings [13]. The inset of Fig. 1 illustrates a typical sample structure. The thickness of the GaN layer in one SL period is denoted by d_{GaN}.

Based on our previous work [5], the GaN layers of the samples were grown with a V/III molar ratio of 100 at 550–600 °C to achieve 2D growth. The growth temperature of GaAs (T_{GaAs}) was varied between 550 and 625 °C while maintaining a V/III ratio of 12. Fig. 1 shows a schematic temperature profile during the growth of a SL. The nominal growth rates were 1.3 Å/s for GaN and 2.7 Å/s for GaAs. The surface morphology of the samples was imaged with an atomic force microscope (AFM) using the contact mode. A high-resolution triple-axis X-ray diffractometer (XRD) with an incident beam monochromator and mirror was used to study the crystalline quality of the superlattices.

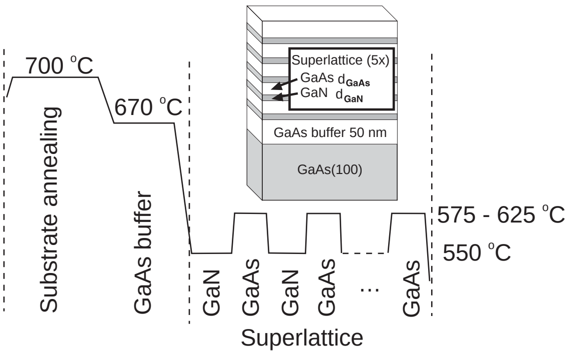

Fig. 1. Schematic temperature profile of the growth of a GaN/GaAs superlattice. The inset shows the structure of a typical sample.

3. Results and discussion

To initiate the study, two-layer samples were grown to optimize the deposition of GaAs (5 nm) on an epitaxial GaN layer (2 nm). When both layers were grown at 550 °C, the surface of the top GaAs layer was rough, shown in Fig. 2(a). The surface was composed of hillocks centered on holes whose diameter was a few tens of nm and whose areal density was in the order of $10^{10}\,\mathrm{cm}^{-2}$. As the growth temperature of both layers was raised, the hillocks gradually changed to two-dimensional (2D) islands (seen in Fig. 2(b)) and coalesced. Next, T_{GaAs} was varied between 575 and 625 °C while growing GaN at the constant 550 °C. This resulted in an atomically flat GaAs surface. With increasing temperature, the growth of GaAs changed from a distinct 2D island growth, shown in Fig. 2(c), to a more step-flow type growth. Simultaneously, the areal density of the holes increased from virtually no holes at 575 °C to $10^{9}\,\mathrm{cm}^{-2}$ at 625 °C. Although their origin remained undetermined, the holes could be caused by dislocations in the GaN layer. Based on these findings, the growth scheme shown in Fig. 1 was chosen for the SLs.

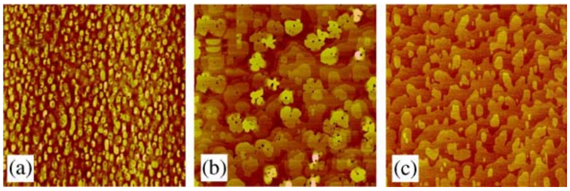

Fig. 2. AFM images (3×3 μm^2) of GaN(2 nm)/GaAs(5 nm) two-layer samples. The GaN/GaAs growth temperatures were (a) 550/550 °C, (b) 600/600 °C, and (c) 550/575 °C, respectively. The vertical scale is 5 nm in (a) and 3 nm in (b) and (c).

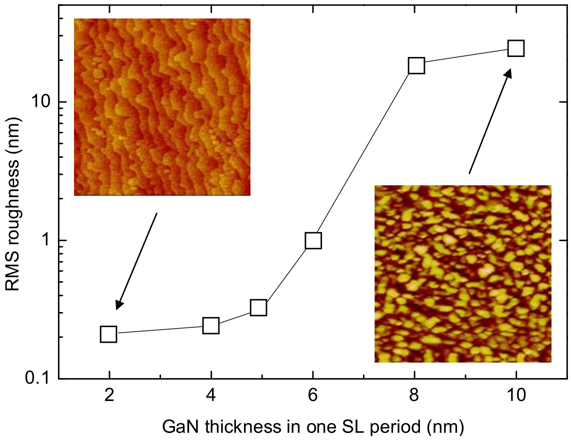

Fig. 3. Root-mean-square (RMS) roughness of SL sample surfaces as a function of GaN layer thickness. The insets show corresponding AFM images from samples with nominally 2 and 10 nm of GaN per period. The vertical scales are 4 and 150 nm, respectively.

The effect of the thickness of the GaN layers on the superlattice was studied by growing a series of 5-period SLs having a nominal period thickness of 30 nm, varying d_{GaN} between 2 and 10 nm. The GaAs layers were grown at 600 °C. Fig. 3 shows the root-mean-square (RMS) roughness of the sample surfaces, calculated from 3×3 μm^2 AFM scans. Up to $d_{\mathrm{GaN}} = 5$ nm, the surfaces are smooth with visible atomic terraces or plateaus and a roughness below 0.4 nm. With $d_{\mathrm{GaN}} = 6$ nm, the surface is rougher, and holes, roughly 100 nm in diameter, with an areal density of 10^9 cm^{-2} were seen (not shown here). As the GaN layer thickness is still increased, the roughness increases rapidly. The insets of Fig. 3 show AFM images of a flat ($d_{\mathrm{GaN}} = 2$ nm) and a rough ($d_{\mathrm{GaN}} = 10$ nm) surface. Note the different vertical scales in the images. The results show that to obtain a SL with a good surface morphology in these growth conditions, the maximum GaN thickness per period is 5 nm.

The morphology of the samples is impaired not only by holes but also by island growth that increases the roughness of the layers. Islanding is typically induced by low growth temperatures where the surface diffusion length of the adatoms is too short to favor step-edge growth. Moreover, heteroepitaxial strain has been shown to induce islanding of e.g., SiGe [14] even at higher growth temperatures. In this study, the growth of the GaAs layers of the SLs is seen to evolve from mainly step-flow growth via layer-by-layer 2D island growth to 3D island growth as d_{GaN} is increased from 2 to 5 nm. This might indicate that the GaN layers apply strain on the epitaxial GaAs, thus suggesting that the GaN is indeed (partially) relaxed.

To verify the influence of d_{GaN} on the quality of the aforementioned SL samples, XRD scans around the GaAs(0 0 4) reflection were measured. Fig. 4 shows the ω–2θ measurement of a GaN/GaAs SL where $d_{\mathrm{GaN}} = 5$ nm. A schematic picture of the measurement setup is shown in the inset. The lower curve is a simulation of a SL with $d_{\mathrm{GaN}} = 5$ nm and $d_{\mathrm{GaAs}} = 24$ nm, giving the best fit with the measurement. Overall, the SL samples

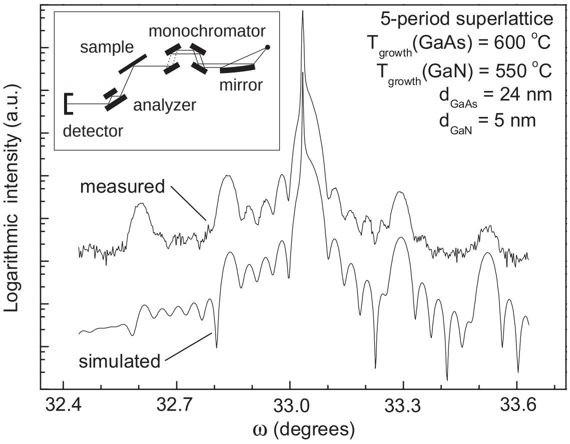

Fig. 4. XRD ω–2θ measurement and simulation of a 5-period GaN(5 nm)/GaAs(24 nm) superlattice. The inset shows a schematic picture of the triple-axis measurement setup.

with $d_{\mathrm{GaN}} \leqslant 6\,\mathrm{nm}$ exhibited similar, clearly identifiable satellite peaks and interference fringes, thus implying good crystal quality and sharp interfaces. However, the samples with $d_{\mathrm{GaN}} = 8$–$10\,\mathrm{nm}$ were deteriorated showing little periodic structure besides the GaAs(0 0 4) reflection. These results are in good agreement with the surface roughness measurements, giving 5–6 nm as the maximum d_{GaN} for high-quality SLs.

Finally, to find out how the growth temperature of the GaAs layers affects the structure, a series of GaN(5 nm)/GaAs(25 nm) SLs were grown with T_{GaAs} ranging from 575 to 625 °C. Fig. 5 shows the X-ray diffraction space mappings and 3×3 $\mu\mathrm{m}^2$ AFM images of the samples. In the space map, measured around the GaAs(0 0 4) reciprocal lattice point, the 2θ–ω axis follows lattice spacing and the $\Delta\omega$ axis follows lattice tilt [15]. Based on the space maps, no significant misorientation between the layers and the substrate is found. As seen in Fig. 5(a), the surface morphology of the SL suffers greatly when grown at 625 °C. However, the diffraction measurement shows a fringe pattern similar to the reference sample (c) grown at 600 °C, indicating reasonably sharp interfaces. Figs. 5(d) and (e) show that as the temperature is decreased from 600 °C, diffuse scattering and morphology indicate degradation of the crystal quality. At 588 and 612 °C (Figs. 5(d) and (b)), a small number of holes can be seen on the surfaces. We note that sample (c) is clear of such holes. Therefore, the results suggest that the optimum T_{GaAs} is around 600 °C to yield both good crystal quality and an acceptable surface morphology of the SL.

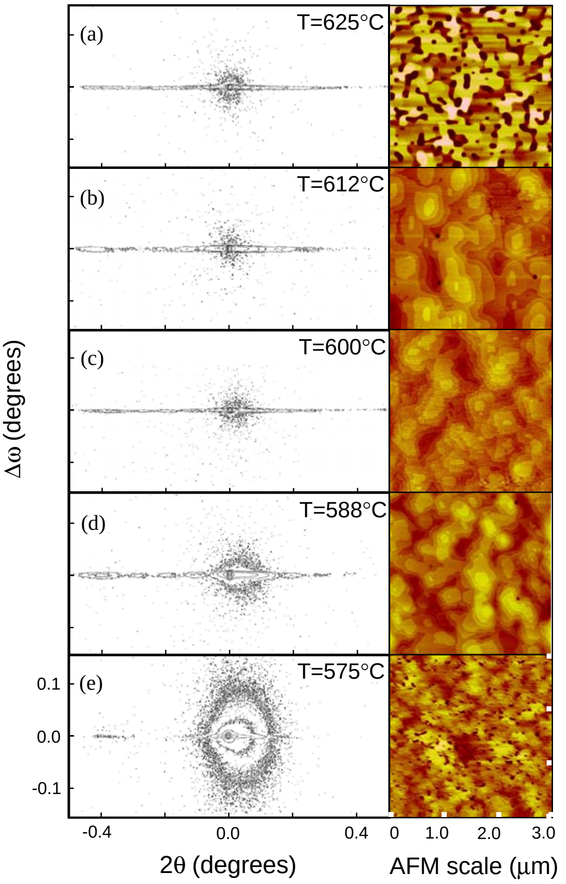

Fig. 5. X-ray diffraction space maps of GaN/GaAs superlattices with 5 nm of GaN per period, measured around the GaAs(0 0 4) reflection. On the right, corresponding 3×3 $\mu\mathrm{m}^2$ atomic force micrographs are shown. The AFM vertical scale is 10 nm in (a) and (e), 5 nm in (b)–(d). GaAs growth temperatures are given in the figures.

4. Conclusions

In summary, we have grown 5-period cubic GaN/GaAs superlattices by MOVPE using DMHy. The SLs with up to 6 nm of GaN in one SL period exhibited strong XRD fringes, implying good crystallinity and sharp interfaces. 3D island growth and holes degraded the morphology of the SLs with more than 5 nm of GaN per period. The formation of holes was found to be strongly dependent on growth temperature and layer thickness. A two-temperature growth scheme, 550 °C for GaN and 600 °C for GaAs, was found to result in 2D layer-by-layer growth and a hole-free morphology. Altogether in light of these findings, the use of MOVPE-grown very-thin GaN layers in GaAs structures seems possible.

References

[1] I. Akasaki, J. Crystal Growth 237–239 (2002) 905–911.

[2] H. Tachibana, T. Ishido, M. Ogawa, M. Funato, S. Fujita, S. Fujita, J. Crystal Growth 196 (1999) 41–46.

[3] J. Wu, H. Yaguchi, K. Onabe, R. Ito, Appl. Phys. Lett. 71 (15) (1997) 2067–2069.

[4] H. Yang, O. Brandt, A. Trampert, K.H. Ploog, Appl. Surf. Sci. 104/105 (1996) 461–467.

[5] J. Sormunen, J. Toivonen, M. Sopanen, H. Lipsanen, Appl. Surf. Sci. 222 (2004) 286–292.

[6] S.A. Ding, S.R. Barman, K. Horn, H. Yang, B. Yang, O. Brandt, K. Ploog, Appl. Phys. Lett. 70 (1997) 2407–2409.

[7] B.K. Agrawal, S. Agrawal, R. Srivastava, Surf. Sci. 424 (1999) 232–243.

[8] G. Dehlinger, U. Gennser, D. Grützmacher, T. Ihn, E. Müller, K. Ensslin, Thin Solid Films 369 (2000) 390–393.

[9] P. Han, M. Sakuraba, Y.-C. Jeong, K. Bock, T. Matsuura, J. Murota, J. Crystal Growth 209 (2000) 315–320.

[10] N. Akiba, F. Matsukura, Y. Ohno, A. Shen, K. Ohtani, T. Sakon, M. Motokawa, H. Ohno, Physica B 256–258 (1998) 561–564.

[11] J.W. Yang, C.J. Sun, Q. Chen, M.Z. Anwar, M. Asif Khan, S.A. Nikishin, G.A. Seryogin, A.V. Osinsky, L. Chernyak, H. Temkin, C. Hu, S. Mahajan, Appl. Phys. Lett. 69 (1996) 3566–3568.

[12] H. Sato, H. Takahashi, A. Watanabe, H. Ota, Appl. Phys. Lett. 68 (1996) 3617–3619.

[13] M.E. Heimbuch, A.L. Holmes Jr., C.M. Reaves, M.P. Mack, S.P. DenBaars, L.A. Coldren, J. Electron Mater. 23 (1994) 87.

[14] G.G. Jernigan, P.E. Thompson, Surf. Sci. 516 (2002) 207–215.

[15] P.F. Fewster, Semicond. Sci. Technol. 8 (1993) 1915–1934.