Title
Structural and morphological characteristics of planar (11(2)over-bar0) a-plane gallium nitride grown by hydride vapor phase epitaxy

Journal
Applied Physics Letters, 83(8)

ISSN
0003-6951

Authors
Haskell, B A
Wu, F
Matsuda, S
et al.

Publication Date
2003-08-01

Peer reviewed

Structural and morphological characteristics of planar $(11\bar{2}0)$ *a*-plane gallium nitride grown by hydride vapor phase epitaxy

B. A. Haskell and F. Wu
Materials Department, University of California, Santa Barbara, California 93106-5050 and NICP/ERATO JST, UCSB Group, University of California, Santa Barbara, California 93106-5050

S. Matsuda
Materials Department, University of California, Santa Barbara, California 93106-5050

M. D. Craven, P. T. Fini, S. P. DenBaars, J. S. Speck,[a] and Shuji Nakamura
Materials Department, University of California, Santa Barbara, California 93106-5050 and NICP/ERATO JST, UCSB Group, University of California, Santa Barbara, California 93106-5050

(Received 26 February 2003; accepted 24 June 2003)

This letter discusses the structural and morphological characteristics of planar, nonpolar $(11\bar{2}0)$ *a*-plane GaN films grown on $(1\bar{1}02)$ *r*-plane sapphire by hydride vapor phase epitaxy. Specular films with thicknesses over 50 μm were grown, eliminating the severely faceted surfaces that have previously been observed for hydride vapor phase epitaxy-grown *a*-plane films. Internal cracks and crack healing, similar to that in *c*-plane GaN films, were observed. Atomic force microscopy revealed nanometer-scale pitting and steps on the film surfaces, with rms roughness of $\sim$2 nm. X-ray diffraction confirmed the films are solely *a*-plane oriented with on-axis $(11\bar{2}0)$ and 30° off-axis $(10\bar{1}0)$ rocking curve peak widths of 1040 and 3000 arcsec, respectively. Transmission electron microscopy revealed a typical basal plane stacking fault density of 4×10^5 cm^{-1}. The dislocation content of the films consisted of predominately edge component ($\mathbf{b}_{\mathrm{edge}}=\pm[0001]$) threading dislocations with a density of 2×10^{10} cm^{-2}, and mixed-character Shockley partial dislocations ($\mathbf{b}=\frac{1}{3}\langle1\bar{1}00\rangle$) with a density of 7×10^9 cm^{-2}. [DOI: 10.1063/1.1604174]

Gallium nitride and its alloys with indium and aluminum nitride have attracted significant attention in recent years due to the successful development of visible and ultraviolet light emitting diodes,[1] blue/violet laser diodes,[2] and high-power electronic devices[3] based on this materials system. While *c*-plane-oriented (Al, In, Ga)N films are the most commonly grown nitrides for device applications, *c*-axis-oriented optoelectronic devices in particular suffer from undesirable spontaneous and piezoelectric polarization effects. Charge separation within quantum wells decreases the electron-hole recombination efficiency and redshifts the emission wavelengths,[4–8] both of which are undesirable in the operation of short-wavelength visible and ultraviolet emitters. These polarization effects can be eliminated by growing devices on alternative orientations of GaN crystals, such as $\{10\bar{1}0\}$ *m*-plane or $\{11\bar{2}0\}$ *a*-plane films, in which the GaN polar axis exists within the planes of the device layers rather than out of them. Plasma-assisted molecular beam epitaxy (MBE)-grown *m* plane[9] and metalorganic chemical vapor deposition (MOCVD)[10] and MBE-grown[11] *a*-plane AlGaN/GaN heterostructures have demonstrated an elimination of polarization fields along their respective growth directions.

As bulk GaN crystals of appreciable size are not available, hydride vapor phase epitaxy (HVPE) has been used to heteroepitaxially grow thick (10–300 μm) *c*-plane GaN films to serve as homoepitaxial substrates for MBE or MOCVD device growth.[12,13] Previous attempts to grow *a*-plane GaN by HVPE, however, have yielded rough and faceted surfaces that are inferior to *c*-plane GaN films for substrate use.[13–15] We report here on the structural and morphological characteristics of planar $(11\bar{2}0)$ GaN films recently grown on *r*-plane sapphire substrates by HVPE.

The *a*-plane GaN films discussed in this letter were grown in a three-zone horizontal directed-flow HVPE system. Commercially available *r*-plane sapphire substrates were loaded directly into the reactor without any *ex situ* cleaning. The *a*-plane GaN films were grown directly on the sapphire substrates without the use of any low-temperature buffer or nucleation layers. GaCl, formed by *in situ* reaction of HCl with 99.99999% Ga, was combined with NH_3 8 mm from the sapphire substrate, with V/III ratios between 9 and 40. Typical growth rates ranged from 15 to 60 μm/h, and films up to 53 μm in thickness were grown at substrate temperatures between 1040 and 1070 °C.

Figure 1(a) shows a Nomarski optical contrast micrograph of a representative *a*-plane GaN film grown by HVPE. This film's surface was characterized by long-range ''flow'' patterns that had peak-to-valley heights of 100–500 nm over 75–500 μm lateral extents, as measured by profilometry. These low-angle surface features scattered light minimally, and the films were specular and optically transparent. Other observed long-range surface features included faint ridges or scales of similar heights but with varying directions over the wafers' surfaces. No correlation was found between these features and the structural properties of the films. In contrast to previously reported *a*-plane GaN surfaces having facets

[a]Author to whom correspondence should be addressed; electronic mail: speck@mrl.ucsb.edu

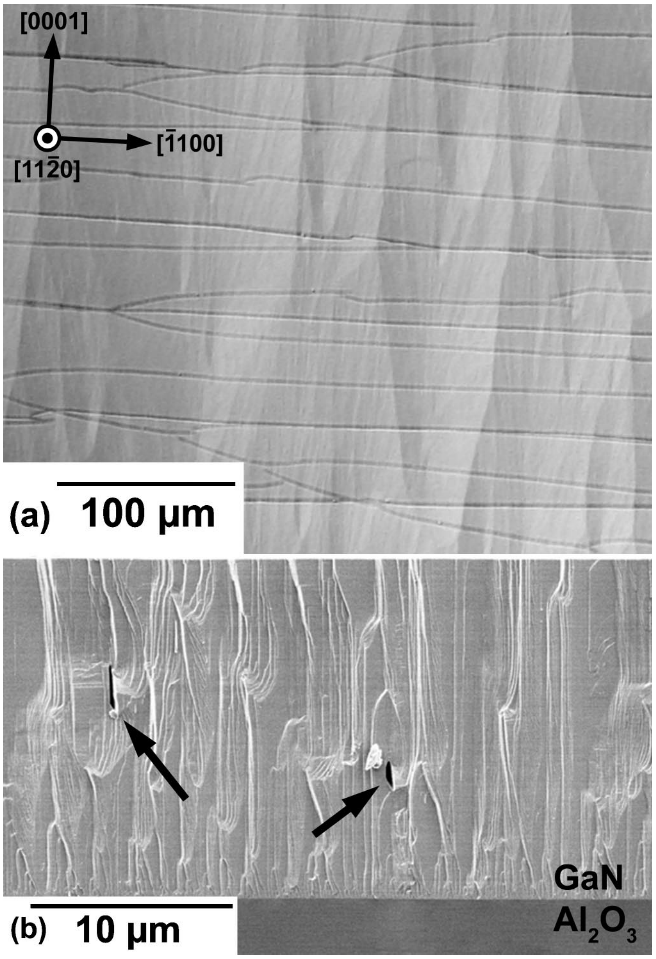

FIG. 1. (a) Nomarski optical contrast micrograph of an *a*-GaN film. (b) Cross-sectional SEM image revealing two internal cracks in an *a*-GaN film.

inclined 30°–50° from the surface normal,[13–16] the angular variation of these surface features was 0.2°–0.8° and cannot be attributed to faceting. Subsurface cracks oriented nearly perpendicular to the GaN *c* axis were observed in this sample, two of which are detailed in the cross-sectional scanning electron micrograph (SEM) in Fig. 1(b). With few exceptions,[16] these internal cracks, which are similar to those observed in *c*-plane GaN films,[17] did not reach the free surface during growth. These cracks result from plastic relief of tensile strain that may be a consequence of grain coalescence.[18] The cracks subsequently heal via lateral overgrowth to reduce surface energy. Detailed discussion of the nature and origin of these cracks will be provided elsewhere.

Figure 2 shows a representative atomic force micrograph (AFM) from an *a*-plane GaN film. Local rms roughness over 2×2 μm sampling areas was typically 0.5–0.8 nm. The rms roughness over 100–400 μm^2 areas remained below 2 nm. The surface was dominated by a high density of 3–7-nm-deep pits with densities ranging from 2×10^9 to 9×10^9 cm^{-2}. These pits likely decorated threading dislocation terminations at the free surface. Additionally, ~1-nm-high steps with a density of $\sim7\times10^4$ cm^{-1} were apparent in the image. In contrast to the long-range surface features, these steps were oriented roughly perpendicular to the GaN *c* axis regardless of flow conditions, and may be related to the presence of basal plane stacking faults in the films.

Structural characteristics of the planar *a*-plane GaN films

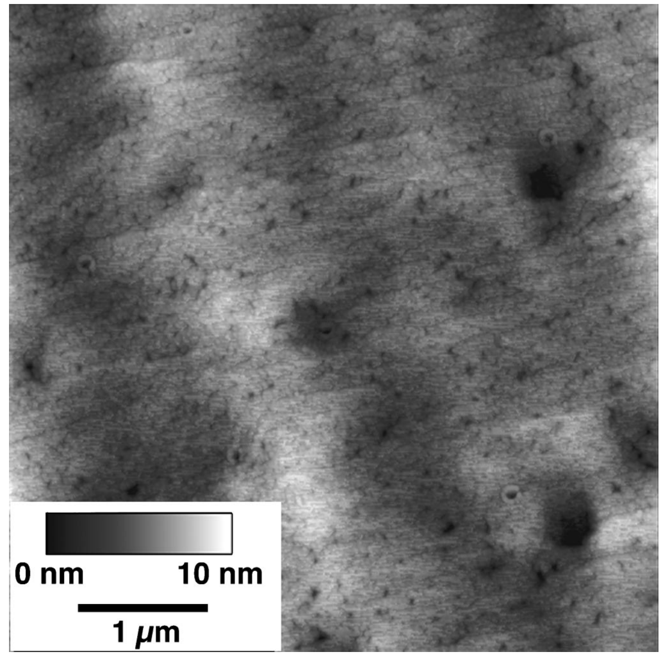

FIG. 2. 5×5 μm AFM image of an *a*-GaN surface.

were evaluated by x-ray diffraction (XRD) and plan-view transmission electron microscopy (TEM). XRD was performed using Cu $K\alpha$ radiation in a Philips MRD Pro four-circle x-ray diffractometer operating in receiving slit mode. The ω-2θ scans of the *a*-GaN films exhibited peaks that were indexed to the *r*-plane sapphire $(1\bar{1}02)$, $(2\bar{2}04)$, and $(3\bar{3}06)$ reflections, and the $(11\bar{2}0)$ GaN reflection. No GaN (0002) reflection was observed, demonstrating that within the detection limits of this technique the films were uniformly *a*-plane oriented. The ω rocking curves were measured on the on-axis $(11\bar{2}0)$ reflection for geometries in which the GaN $[\bar{1}100]$ and [0001] directions were in a coplanar geometry. Typical full widths at half maximum (FWHM) for the $(11\bar{2}0)$ reflection in these geometries were generally comparable at 1040–1045 arcsec. The 30° off-axis $(10\bar{1}0)$ reflection was measured by tilting the samples relative to the scattering plane (skew geometry), yielding a FWHM on the order of 3000 arcsec. The on-axis peak width was comparable to that observed for planar MOCVD-grown *a*-plane GaN films,[19] while the off-axis peak width was roughly twice as large, indicating a more defined cell structure and higher mosaic content in the HVPE-grown film.[20]

Figure 3 shows plan-view transmission electron micrographs of an *a*-plane GaN film. Figure 3(a) was imaged under the $\mathbf{g}=0002$ diffraction condition, revealing threading dislocations having a Burgers vector component parallel to the GaN [0001] direction. Thus, these are edge component dislocations. The *c*-component dislocation density ranged from 9×10^9 to 2×10^{10} cm^{-2} in these samples, comparable to AFM pit density measurements and TEM of MOCVD-grown *a*-plane GaN films.[19] The TEM image in Fig. 3(b), taken under the $\mathbf{g}=1\bar{1}00$ diffraction condition, shows a stacking fault density of $\sim4\times10^5$ cm^{-1}, again comparable with the 3.8×10^5 cm^{-1} observed for planar MOCVD-grown *a*-plane GaN films.[19] These basal-plane stacking faults are likely related to the presence of exposed nitrogen-face $(000\bar{1})$ surfaces during the early stages of growth. Additional

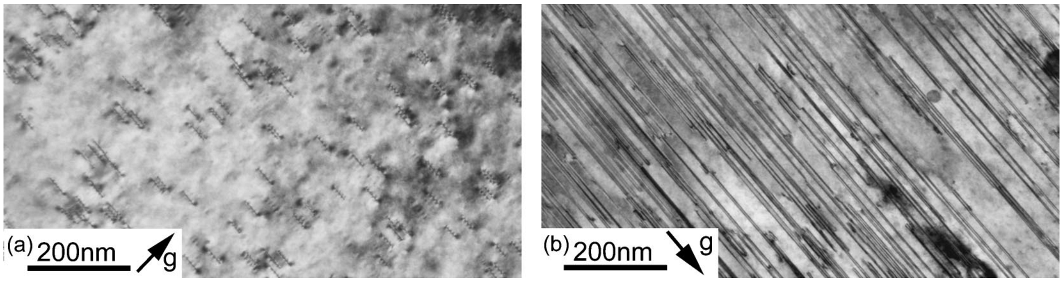

FIG. 3. Plan-view TEM images as observed with (a) $\mathbf{g}=0002$ to observe dislocations having a nonzero [0001] component, and (b) $\mathbf{g}=1\bar{1}00$ to observe stacking faults parallel to the (0001) plane.

imaging with varying sample tilt in the $\mathbf{g}=1\bar{1}00$ diffraction condition revealed $\sim 7\times 10^9$ cm^{-2} Shockley partial dislocations having Burgers vectors $\mathbf{b}=\frac{1}{3}\langle 1\bar{1}00\rangle$.

In conclusion, highly planar nonpolar *a*-plane GaN has been grown by HVPE. The resulting films are specular with limited internal cracking being apparent by Nomarski optical microscopy and SEM. The surfaces are virtually free of macroscopic defects while possessing nanometer-scale steps and pits decorating threading dislocation terminations. These films are structurally and morphologically comparable to MOCVD-grown *a*-plane GaN films and are suitable for use as template layers for polarization-free device fabrication. With incremental improvement in structural and morphological characteristics, the development of nonpolar GaN substrates can now be realized, potentially yielding high-efficiency visible and ultraviolet optoelectronic devices.

This work made use of the MRL Central Facilities supported by the National Science Foundation under Award No. DMR00-80034.

[1] T. Nishida and N. Kobayashi, Phys. Status Solidi A **188**, 113 (2001).
[2] S. Nakamura, G. Fasol, and S. J. Pearton, *The Blue Laser Diode* (Springer, New York, 2000).
[3] L. F. Eastman and U. K. Mishra, IEEE Spectrum **39**, 28 (2002).
[4] T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, I. Akasaki, Y. Kaneko, S. Nakagawa, Y. Yamaoka, and N. Yamada, Appl. Phys. Lett. **73**, 1691 (1998).
[5] I. Jin Seo, H. Kollmer, J. Off, A. Sohmer, F. Scholz, and A. Hangleiter, Phys. Rev. B **57**, R9435 (1998).
[6] R. Langer, J. Simon, V. Ortiz, N. T. Pelekanos, A. Barski, R. Andre, and M. Godlewski, Appl. Phys. Lett. **74**, 3827 (1999).
[7] P. Lefebvre, J. Allegre, B. Gil, H. Mathieu, N. Grandjean, M. Leroux, J. Massies, and P. Bigenwald, Phys. Rev. B **59**, 15363 (1999).
[8] P. Lefebvre, A. Morel, M. Gallart, T. Taliercio, J. Allegre, B. Gil, H. Mathieu, B. Damilano, N. Grandjean, and J. Massies, Appl. Phys. Lett. **78**, 1252 (2001).
[9] P. Waltereit, O. Brandt, A. Trampert, H. T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, and K. H. Ploog, Nature (London) **406**, 865 (2000).
[10] M. D. Craven, P. Waltereit, F. Wu, J. S. Speck, and S. P. DenBaars, Jpn. J. Appl. Phys., Part 2 **42**, L235 (2003).
[11] H. M. Ng, Appl. Phys. Lett. **80**, 4369 (2002).
[12] K. Motoki, T. Okahisa, N. Matsumoto, M. Matsushima, H. Kimura, H. Kasai, K. Takemoto, K. Uematsu, T. Hirano, M. Nakayama, S. Nakahata, M. Ueno, D. Hara, Y. Kumagai, A. Koukitu, and H. Seki, Jpn. J. Appl. Phys., Part 2 **40**, L140 (2001).
[13] T. Paskova, P. P. Paskov, V. Darakchieva, S. Tungasmita, J. Birch, and B. Monemar, Phys. Status Solidi A **183**, 197 (2001).
[14] A. Shintani and S. Minagawa, J. Electrochem. Soc. **123**, 1575 (1976).
[15] R. Molnar, in *Semiconductors and Semimetals Vol. 57: Gallium Nitride*, edited by J. I. Pankove and T. D. Moustakas (Elsevier, New York), pp. 1–31.
[16] M. Sano and M. Aoki, Jpn. J. Appl. Phys. **15**, 1943 (1976).
[17] E. V. Etzkorn and D. R. Clarke, J. Appl. Phys. **89**, 1025 (2001).
[18] T. Böttcher, S. Einfeldt, S. Figge, R. Chierchia, H. Heinke, D. Hommel, and J. S. Speck, Appl. Phys. Lett. **78**, 1976 (2001).
[19] M. D. Craven, S. H. Lim, F. Wu, J. S. Speck, and S. P. DenBaars, Appl. Phys. Lett. **81**, 469 (2002).
[20] B. Heying, X. H. Wu, A. S. Keller, Y. Li, D. Kapolnek, B. P. Keller, S. P. DenBaars, and J. S. Speck, Appl. Phys. Lett. **68**, 643 (1996).