APPLIED PHYSICS LETTERS **89**, 103106 (2006)

Very high quality AlN grown on (0001) sapphire by metal-organic vapor phase epitaxy

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(Received 16 May 2006; accepted 24 July 2006; published online 6 September 2006)

Very high quality AlN epitaxially grown on (0001) sapphire by metal-organic vapor phase epitaxy is investigated by atomic force microscopy, x-ray diffraction, and photospectrometry. A clear and continuously linear step-flow pattern with sawtooth shaped terrace edges is observed in atomic force microscopic images. Triple-axis x-ray rocking curves show a full width at half maximum of 11.5 and 14.5 arc sec for the (002) and (004) reflections, respectively. KOH etching reveals an etch-pit density of 2×10^7 cm^{-2}, as deduced from atomic force microscopy measurements. The optical transmission spectrum shows a sharp absorption edge with a band gap energy of 6.10 eV. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2345256]

Aluminum nitride (AlN) has generated much interest due to its unique properties such as its very wide and direct band gap and high thermal conductivity. High quality AlN epitaxial layers are needed in AlGaN based ultraviolet (UV) light-emitting diodes (LEDs), which have great potential for applications such as fluorescence-based biological agent detection, water purification, sterilization and decontamination, non-line-of-sight communications, and thin-film curing.[1] At the same time, deep UV photodetectors need extremely high quality AlN with low dislocation densities for the reduction of dark current. Many efforts have been made in order to improve the crystalline quality of AlN and to decrease the dislocation density.[2–9] Nevertheless, the current crystalline quality of epitaxial AlN must be improved particularly for UV LEDs and photodetectors.

In this letter, we report on the growth of AlN layers on (0001) sapphire using the low temperature (LT) AlN nucleation scheme.[10] The material quality of the AlN epitaxial layers grown on sapphire is determined by atomic force microscopy (AFM), x-ray diffraction (XRD), etch-pit density (EPD) measurement, scanning electron microscopy (SEM), and photospectrometry. The results demonstrate that very high quality AlN layers can be grown on sapphire.

The AlN epitaxial layers are grown using an Aixtron 200/4-RF S low pressure metal-organic vapor phase epitaxy (MOVPE) system with a single-wafer horizontal-flow geometry and with radio-frequency heating. Growth is initiated by a 7-nm-thick LT AlN nucleation layer grown at 840 °C of reactor temperature and a reactor pressure of 50 mbars followed by a 1-μm-thick high temperature (HT) AlN layer grown at 1215 °C and 25 mbars. The molar V/III ratios of the LT and HT AlN layers are 6397 and 483, respectively. The surface quality of the HT AlN layer, assessed by AFM, is shown in Fig. 1. A very clear and continuously linear step-flow pattern is observed on a 5×5 μm^2 size image, which indicates very high crystalline quality.[5,6] On the 2×2 μm^2 size image, shown in Fig. 1(b), the equidistant terrace displays sawtooth shaped edges, which is very similar with what we observed on AlN epitaxial layers grown on AlN bulk substrates. Thus, such a step-flow pattern with sawtooth shaped terrace edges shows that the epitaxial layer is of very high crystalline quality. The root mean square roughnesses of Figs. 1(a) and 1(b) are 0.188 and 0.181 nm, respectively.

X-ray rocking curves are measured on the AlN layer with triple-axis optics and double-axis optics by using a PANalytical PW3040/60 X'Pert Pro system. Figure 2 shows the (002) and (004) rocking curves obtained with triple-axis optics. The full widths at half maximum (FWHMs) of the (002) and (004) peaks are 11.5 and 14.5 arc sec, respectively. The FWHMs of the (002) and (004) peaks measured by double-axis optics are 32.4 and 54.4 arc sec, respectively. Our FWHM values are even comparable to the best AlN *bulk* materials, which is about 10 arc sec, also measured by triple-axis optics.[11] The asymmetric (104) peak rocking-curve scan is measured by using double-axis optics and shows the FWHM of 155 arc sec.

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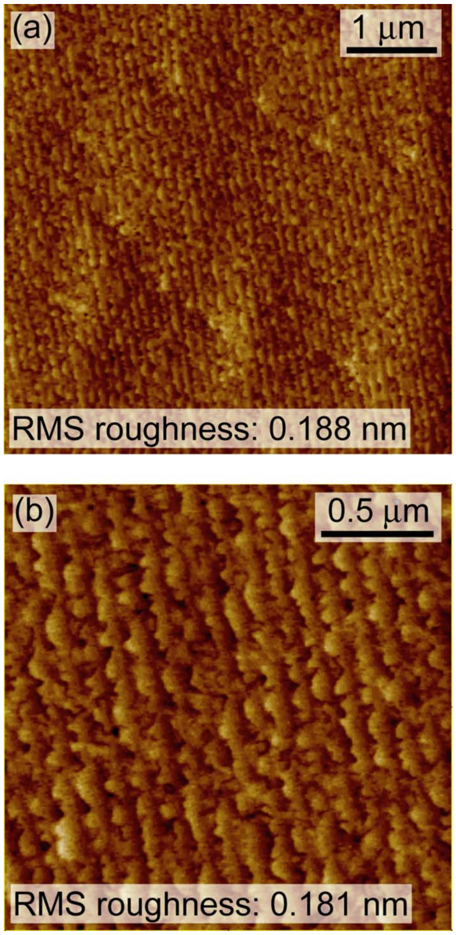

FIG. 1. (Color online) AFM images of HT AlN surface grown on (0001) sapphire. (a) 5×5 μm^2 image and (b) 2×2 μm^2 image.

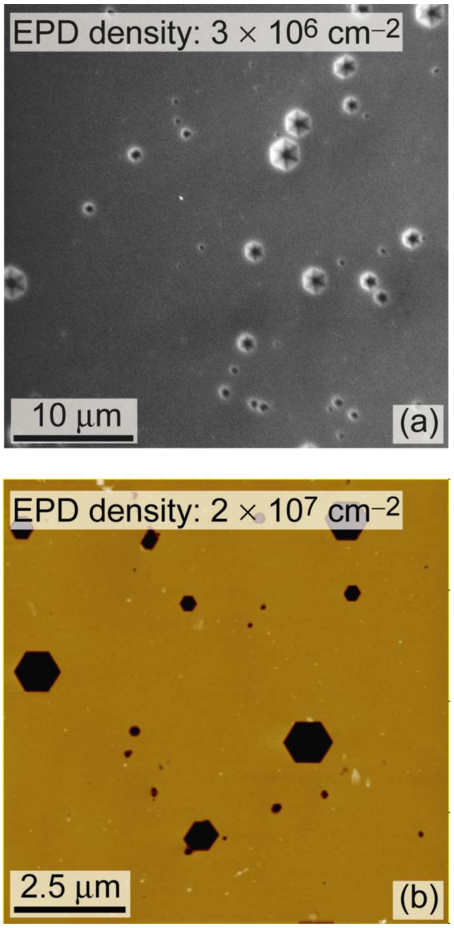

FIG. 3. (Color online) (a) SEM image and (b) AFM image of KOH-etched AlN epitaxial layer used to evaluate the etch-pit density.

In order to assess the surface quality, an etch-pit density study is performed. The AlN sample is etched for 45 min in a 50% KOH solution heated to 70 °C. The etched surface is measured by SEM and AFM, and the results are shown in Fig. 3. Before the SEM measurement, an 8-nm-thick Au film is e-beam deposited to work as a conductive layer. An etch-pit density of 3×10^6 cm^{-2} is obtained from the SEM measurement, as shown in Fig. 3(a), and 2×10^7 cm^{-2} from the AFM measurement, as shown in Fig. 3(b). The EPDs measured by AFM are higher since the spatial resolution of AFM is better, and submicron-scale etch pits can be easily observed. By comparing the AFM and SEM results, we find that some very small etch pits are not detectable by SEM. Hence, AFM will be the better technology to evaluate EPD density.

The optical transmittance spectrum of the AlN epitaxial layer on sapphire is measured using a Jasco V570 spectrophotometer. The spectrum, shown in Fig. 4, displays a sharp absorption edge and a band gap energy of 6.10 eV is evaluated based on the absorption coefficient calculation,[12] as shown in the inset of the figure. The band gap energy of 6.10 eV is in excellent agreement with the literature value of 6.077 eV.[13]

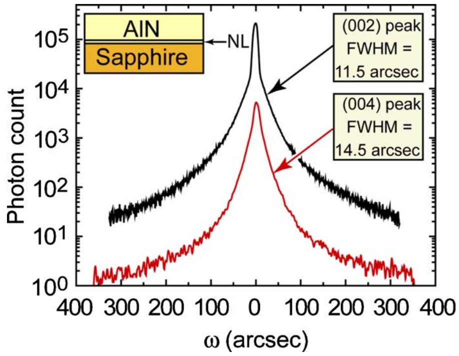

FIG. 2. (Color online) X-ray rocking curves of AlN (002) and (004) peaks measured by triple-axis optics. FWHMs of 11.5 and 14.5 arc sec are obtained for the (002) and (004) reflections, respectively.

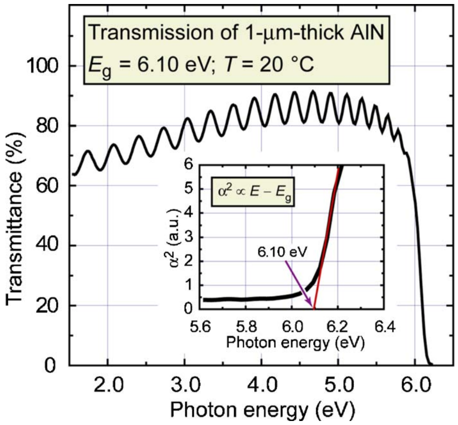

FIG. 4. (Color online) Optical transmittance spectrum of AlN grown on (0001) sapphire. A band gap energy of 6.096 eV is obtained.

High quality AlN is very difficult to grow due to a narrow growth window, strong parasitic reactions at elevated temperatures, and the large number of parameters, such as growth temperature, trimethylaluminum (TMAl) flow, ammonia flow, and growth time of the nucleation layer, which need to be controlled simultaneously with high accuracy. We also find that low quality nucleation layers cause micrometer-sized openings on the HT AlN surface, consistent with results reported in literature.[5,6] In the present study, it is also found that very different results were obtained from two growths with the same V/III ratio, but different ammonia and TMAl flows. HT AlN growth prefers relatively low V/III ratio. In our case, a V/III ratio of 483 is used. This can be explained by assuming that a low V/III ratio reduces parasitic reactions occurring at very high temperature.

In conclusion, very high quality AlN epitaxial layers grown on sapphire (0001) by MOVPE are presented. A very clear and continuously linear step-flow pattern is observed on a $5 \times 5\ \mu\text{m}^2$ size AFM image. A sawtooth shaped terrace edge of the step-flow pattern indicates a very high crystalline quality. FWHM of XRD rocking-curve scans are as narrow as 11.5 arc sec and thus comparable with that of the best AlN bulk material. An EPD of $2 \times 10^7\ \text{cm}^{-2}$ measured by AFM is obtained on KOH-etched AlN surface. The transmittance spectrum shows a very sharp absorption edge and band gap energy of 6.10 eV.

Support through the Crystal IS, DOE, ARO, NSF, Samsung Advanced Institute of Technology, Sandia National Laboratories, and New York Sate is gratefully acknowledged. The authors would like to acknowledge contributions from Thomas Gessmann and useful discussions with Leo J. Schowalter of Crystal IS and Jan-Yves Clames of Aixtron Inc.

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