Journal of Crystal Growth 414 (2015) 76–80

Contents lists available at ScienceDirect

Journal of Crystal Growth

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

Temperature dependence of the crystalline quality of AlN layer grown on sapphire substrates by metalorganic chemical vapor deposition

Xiao-Hang Li [a,*], Yong O. Wei [b], Shuo Wang [b], Hongen Xie [b], Tsung-Ting Kao [a], Md. Mahbub Satter [a], Shyh-Chiang Shen [a], P. Douglas Yoder [a], Theeradetch Detchprohm [a], Russell D. Dupuis [a,c,**], Alec M. Fischer [b], Fernando A. Ponce [b]

[a] *Center for Compound Semiconductors and School of Electrical and Computer Engineering, Georgia Institute of Technology, 777 Atlantic Dr. NW, Atlanta, GA 30332, USA*
[b] *Department of Physics, Arizona State University, Tempe, AZ 85287, USA*
[c] *School of Materials Science and Engineering, Georgia Institute of Technology, 777 Atlantic Dr. NW, Atlanta, GA 30332, USA*

ARTICLE INFO

Article history:
Received 15 July 2014
Received in revised form
23 September 2014
Accepted 5 October 2014
Available online 14 October 2014

Keywords:
A1. Characterization
A3. Metalorganic chemical vapor deposition
B1. Nitrides
B2. Semiconducting aluminum compounds

ABSTRACT

We studied temperature dependence of crystalline quality of AlN layers at 1050–1250 °C with a fine increment step of around 18 °C. The AlN layers were grown on *c*-plane sapphire substrates by metalorganic chemical vapor deposition (MOCVD) and characterized by X-ray diffraction (XRD) ω-scans and atomic force microscopy (AFM). At 1050–1068 °C, the templates exhibited poor quality with surface pits and higher XRD (002) and (102) full-width at half-maximum (FWHM) because of insufficient Al atom mobility. At 1086 °C, the surface became smooth suggesting sufficient Al atom mobility. Above 1086 °C, the (102) FWHM and thus edge dislocation density increased with temperatures which may be attributed to the shorter growth mode transition from three-dimension (3D) to two-dimension (2D). Above 1212 °C, surface macro-steps were formed due to the longer diffusion length of Al atoms than the expected step terrace width. The edge dislocation density increased rapidly above 1212 °C, indicating this temperature may be a threshold above which the impact of the transition from 3D to 2D is more significant. The (002) FWHM and thus screw dislocation density were insensitive to the temperature change. This study suggests that high-quality AlN/sapphire templates may be potentially achieved at temperatures as low as 1086 °C which is accessible by most of the III-nitride MOCVD systems.

1. Introduction

Deep-ultraviolet (DUV) emitters based on III-nitrides including light-emitting diodes (LEDs) and laser diodes (LDs) can be applied to important areas such as disinfection and digital data storage. Although bulk AlN substrates are ideal for the growth of DUV emitters due to their low-dislocation density as well as reduced lattice and thermal mismatch with high-Al content III-nitrides [1], the application is constrained by limited supply, high cost, DUV absorption, and small substrate size. Thus commercialization of the III-nitride DUV emitters still largely relies on the use of AlN hetero-epitaxial templates grown on the vastly-available, low-cost, DUV-transparent, and larger foreign substrates especially *c*-plane sapphire substrates. However, the large lattice and thermal mismatch between AlN and sapphire leads to high dislocation density. As performance of III-nitride DUV emitters is very sensitive to the dislocation-related non-radiative recombination centers [2,3], it is crucial to reduce the dislocation density of AlN templates for the devices grown and fabricated thereon.

One of the common approaches to reducing the dislocation density is epitaxial lateral overgrowth (ELO) where AlN layers are re-grown on patterned seeding AlN templates [4–7]. However, because the ELO approach involves cleanroom fabrication like lithography and etching as well as a regrowth process of the several μm thick layer to coalesce and form a flat layer over the patterned templates, it is associated with much more additional cost and processing time, uneven surfaces, and growth complexity. Another common approach, the use of pulsed atomic layer epitaxy (PALE), has been implemented where supplies of N and/or Al sources are supplied in a pulse mode to allow Al atoms additional time to mobilize on the epitaxial surface [3,8,9]. In some studies,

* Corresponding author. Tel.: +1 404 894 1722.
** Corresponding author at: Center for Compound Semiconductors and School of Electrical and Computer Engineering, Georgia Institute of Technology, 777 Atlantic Dr. NW, Atlanta, GA 30332, USA. Tel.: +1 404 895 6094.
E-mail addresses: xli@gatech.edu (X.-H. Li), dupuis@gatech.edu (R.D. Dupuis).

http://dx.doi.org/10.1016/j.jcrysgro.2014.10.007
0022-0248/

the ELO and PALE were both employed to accelerate the coalescence over the patterned templates [5,7].

In addition to the ELO and PALE, high-temperature growth above 1200 °C has been employed independently or jointly with the ELO and PALE to achieve low dislocation density and smooth surface morphology by metalorganic chemical vapor deposition (MOCVD), in that the mobility of Al atoms on the epitaxial surface is enhanced at high temperature [10–17]. However, there are some concerns regarding high-temperature growth. Not only does it require a special reactor configuration and/or part coating to reach high temperature, but it can also cause considerable thermal stress and cracks in the heteroepitaxial film due to the large thermal expansion mismatch between AlN and sapphire [18]. In addition, the serious wafer bowing of AlN templates at high temperature can also deteriorate wafer uniformity in terms of the template thickness and the composition of layers grown heteroepitaxially on the template [19,20]. To overcome these issues, there have been some attempts to grow AlN layers on sapphire [21] and SiC [22] substrates below 1200 °C. However, surface of these AlN templates was found to possess a high density of point defects [21,22]. In other words, there have been very few successful studies of growing high-quality planar AlN templates on the sapphire substrates below 1200 °C to our knowledge. In addition, temperature-dependent experiments were carried out previously to grow AlN templates at 1100–1500 °C [16,17,21]. However, the studies were performed with a large increment step of 100 °C. Hence it was not possible to find out variation of crystalline quality within a smaller range of temperature and thus potential optimum temperatures therein.

In this work, we carried out a temperature-dependent experiment to investigate the temperature influence on the crystalline quality of two-layer AlN templates. The template temperatures T_t ranged from 1050 to 1250 °C with a fine increment step of around 18 °C. The goal was to identify trends of crystalline quality variation and a proper temperature range which could lead to sufficient Al mobility for the growth of high-quality AlN/sapphire templates. Details of growth process and characterization are presented hereafter. Neither the ELO nor PALE approach was used in this study. The AlN crystalline quality was characterized by X-ray diffraction (XRD) and atomic force microscopy (AFM).

2. Temperature dependence of crystalline quality of AlN template

2.1. Experimental

To investigate the temperature dependence of the AlN template quality, 12 crack-free AlN templates with a simple two-layer structure were grown by MOCVD at various template temperatures T_t on two-inch diameter c-plane sapphire substrates with an offcut angle of 0.2° toward the m-plane. This offcut angle is expected to lead to an atomic step terrace width of ~89 nm. The MOCVD system used in this study was an AIXTRON 3 × 2 in. close-coupled-showerhead (CCS) system. The emissivity-corrected surface temperature was measured by a dual-wavelength multiple-point pyrometric profiling system. Trimethylaluminum (TMAl) and ammonia (NH_3) were used as precursors and hydrogen (H_2) as carrier gas. Prior to the AlN growth, the susceptor and sapphire substrates were baked at 1100 °C for 5 min in a H_2 ambient. A dose of 0.57 μmol of TMAl was deposited at 930 °C after the bake to pre-condition the sapphire surface for Al-polar AlN layers [23]. It is noted that the dose of TMAl can vary depending on the reactor design. For instance, our similar 6 × 2 in. CCS MOCVD system does not need any TMAl pre-conditioning to grow Al-polar AlN layers on the sapphire substrate. The lack of a requirement for TMAl pre-conditioning was also observed by Reentila et al. using an 11 × 2 in. planetary MOCVD system [11]. However, with insufficient TMAl pre-conditioning in our 3 × 2 in. CCS MOCVD reactor, the surface of the AlN/sapphire template becomes rough and hazy, which is attributed to the mixed polarity AlN [23,24], as exemplarily exhibited

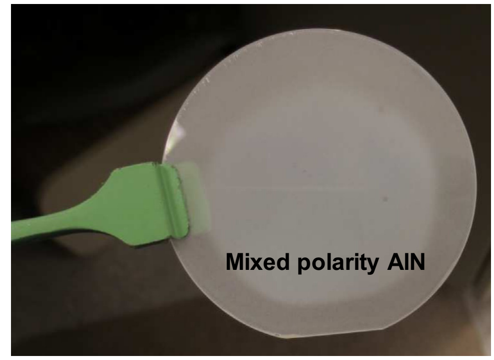

Fig. 1. Photograph of the AlN/sapphire template grown by the AIXTRON 3 × 2 in. CCS MOCVD system with insufficient dose of TMAl pre-conditioning showing rough and hazy surface because of mixed polarity.

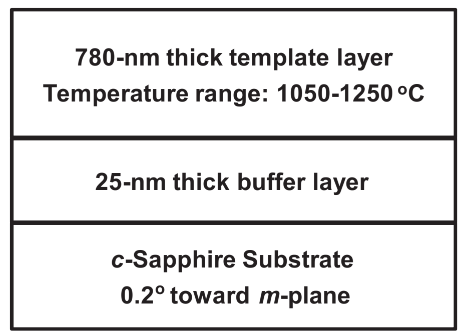

Fig. 2. Cross-sectional schematic diagram of the two-layer AlN templates structures grown on the (0001) sapphire substrates with different template layer temperatures T_t at 1050–1250 °C.

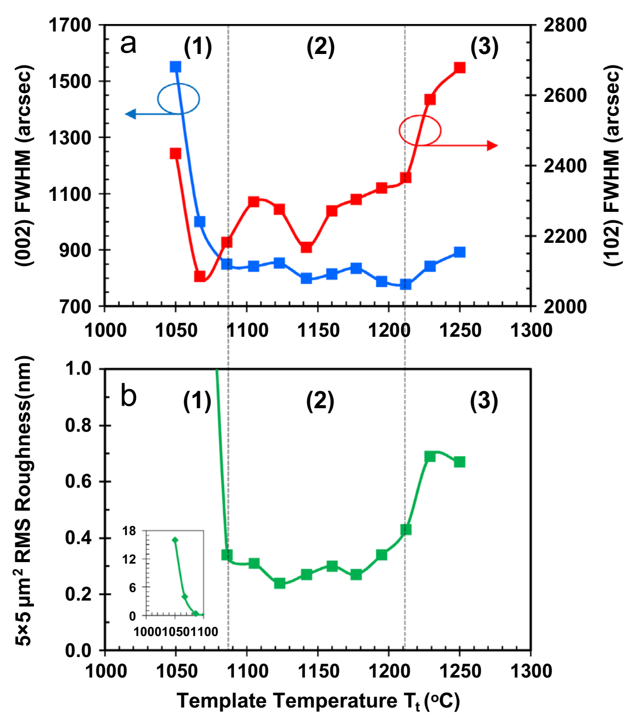

Fig. 3. (a) FWHM's of XRD (002) and (102) ω-scans, and (b) 5 × 5 μm² RMS roughness extracted from Fig. 4 of the two-layer AlN/sapphire templates as a function of temperatures T_t.