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An aberration-corrected STEM study of structural defects in epitaxial GaN thin films grown by ion beam assisted MBE

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ABSTRACT

Ion-beam assisted molecular-beam epitaxy was used for direct growth of epitaxial GaN thin films on super-polished 6H–SiC(0001) substrates. The GaN films with different film thicknesses were studied using reflection high energy electron diffraction, X-ray diffraction, cathodoluminescence and primarily aberration-corrected scanning transmission electron microscopy techniques. Special attention was devoted to the microstructural characterization of GaN thin films and the GaN–SiC interface on the atomic scale. The results show a variety of defect types in the GaN thin films and at the GaN–SiC interface. A high crystalline quality of the produced hexagonal GaN thin films was demonstrated. The gained results are discussed.

1. Introduction

Gallium nitride (GaN) is a "III–V" semiconducting material with a wide bandgap of 3.45 eV and a direct bandgap (Monemar, 1974). Due to this property, it has many applications such as optoelectronic devices, high-mobility field effect transistors and sensor technologies (Nakamura et al., 2000; Pankove, 1973; Takahashi et al., 2007). The efficiency of light emitting devices is directly correlated with the microstructure of the material from which it is made of. Thus, the research in improvement of the material is an important issue. Since a variety of defects influences the emission efficiency of GaN based devices (Yam et al., 2011), a production process allowing the growth of GaN thin films with a low density of defects is desirable. The critical issue in optimization of the epitaxial growth is the lattice mismatch between available substrate materials and the GaN thin films.

Various single crystalline substrate materials are used for deposition of GaN thin films. Widely used substrate materials for GaN epitaxial growth include c-plane sapphire (16% lattice mismatch), r-plane sapphire (1.3% and 16% lattice mismatch), GaAs (20% lattice mismatch), Si (17% lattice mismatch), ZnO (1.8% lattice mismatch) and 6H–silicon carbide (3.5% lattice mismatch). However, regarding the lattice and thermal mismatch, the 6H–SiC(0001) substrate is best suited for GaN thin film heteroepitaxial growth. The lattice mismatch between the GaN film and the substrate is therefore considered as a source of high density of misfit dislocations which propagate into the GaN layer forming threading dislocations. These dislocations can deteriorate physical properties of the GaN thin films. One possibility to overcome this problem is the use of AlN or AlGaN buffer layers (Yoshida et al., 1983) or the growth of thick GaN films (several tens of micrometers) in order to overgrow these defects (Waltereit et al., 2002). Both ways are commonly used nowadays. However, for achieving GaN/SiC heterojunctions with the desired properties for high power and high temperature devices, direct growth of GaN on SiC is preferable.

Today, heteroepitaxial growth of GaN thin films can be achieved by metal-organic chemical vapor deposition (MOCVD) (Matloubian and Gershenzon, 1985), pulsed laser deposition (Vispute et al., 1997), hydride vapor phase epitaxy (HVPE) (Haskell et al., 2003), molecular beam epitaxy (MBE) and time-dependent variations of MBE like migration-enhanced epitaxy (Wong et al., 2011; Kawaharazuka et al., 2010), metal-modulated epitaxy (Moseley

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et al., 2009) and rf-plasma-assisted MBE with pulsed N/Ga atomic deposition (Mei et al., 2008). These variations of MBE techniques for GaN growth have the advantage of higher Ga adatom mobility under Ga-rich growth conditions. As a result, GaN films with low defect densities can be achieved. An alternative method for synthesis of GaN thin films is ion-beam assisted MBE (IBA-MBE) that simultaneously supplies low energy by hyperthermal nitrogen ion irradiation during GaN growth (Gerlach et al., 2000b; Neumann et al., 2012). With this technique, synthesis of GaN thin films with a low density of extended defects compared to the GaN thin films grown by conventional MBE is possible (Sienz et al., 2004a). In addition, this method allows the growth of GaN thin films on SiC substrates without using any kind of buffer layers. However, microstructural properties of these thin films have not been investigated in detail up to now. For optimization of growth conditions and for identifying an application potential of IBA-MBE for GaN production, a detailed characterization of defects and real structure of these films is indispensable.

GaN exists in two different crystalline structures. These structures are hexagonal wurtzite (w-GaN) and cubic zinc blende (z-GaN). The w-GaN is the stable structure whereas the z-GaN is the metastable structure. Both GaN structures can be present in deposited GaN thin films. However, the formation mechanism of z-GaN in GaN thin films is still under discussion.

In this work, the microstructure of GaN thin films deposited by IBA-MBE directly on 6H–SiC substrates without using any kind of buffer layers is investigated. The special focus of this work is on the detailed structural characterization of the GaN thin films grown on 6H–SiC (0001) substrates by aberration-corrected scanning transmission electron microscopy (Cs-corrected STEM). This work is aimed to gain new information on the formation mechanism of GaN thin films by IBA-MBE.

2. Materials and methods

GaN thin films were deposited by IBA-MBE in an ultra-high vacuum system (base pressure $<5 \times 10^{-7}$ Pa) (Neumann et al., 2012). As substrate material, super-polished (0001)-oriented 6H–SiC was chosen as the most suitable material due to its small lattice mismatch of 3.5% and quite similar thermal and mechanical properties (Lin et al., 1993). The substrate temperature was kept constant at 700 °C. This temperature was found to be the optimal temperature for growing of dense GaN thin films with high crystalline quality. The nitrogen ion to gallium atom arrival ratio (I/A ratio) was 1.3 during the entire deposition process (Gerlach et al., 2000a; Neumann et al., 2012). The gallium atoms were provided by a conventional effusion cell and the nitrogen ions by a hollow anode ion source (Anders et al., 1996). The ion source highly efficiently ionizes the passing N_2 gas in a constricted DC glow discharge, resulting in N^+ and N_2^+ ions accelerated towards the sample surface. These ions have a maximum kinetic energy of 25 eV, which is low enough to avoid point defect creation below the surface as well as sputter processes at the sample surface. The nitrogen ion flux was constant at a value of 1.6×10^{14} ions cm^{-2} s^{-1} during the deposition. The hollow anode ion source was operated with constant nitrogen gas flow of 12 sccm leading to a working pressure of 7.5×10^{-2} Pa in the deposition chamber. The Ga effusion cell was operated at 1010 °C resulting in a Ga flux of 1.2×10^{14} atoms cm^{-2} s^{-1}. For the results presented in this paper samples were produced with the same growth conditions but different deposition times of 32 min (sample A) and 150 min (sample B). Eventually, in order to saturate surplus Ga atoms, accumulating at the film surface in form of small Ga droplets during the growth process, a post-ion beam nitridation step with closed Ga source shutter was performed, until the RHEED pattern yielded highest intensity indicating accomplished saturation (Gerlach et al., 2012).

Since the deposition process takes place under Ga-rich growth conditions, a 2D-growth mechanism is preferred (Heying et al., 2000; Okumura et al., 1998). In the present work, the deposition process was monitored *in situ* by reflection high energy electron diffraction (RHEED) with an electron acceleration voltage of 30 kV. The electron beam was in the $[2\bar{1}\bar{1}0]$ azimuth of hexagonal GaN and of the SiC substrate.

The evaluation of the crystalline quality of the GaN thin films was performed by *ex situ* X-ray diffraction (XRD) for evaluating the average tilt and twist rotation angles of the crystallites by means of the rocking curve method. For the rocking curve (ω-scan) measurements a high-resolution diffractometer using a collimated and monochromatic Cu-Kα_1 beam with a wavelength of 0.15406 nm was applied. The diffractometer with a four-axis goniometer allows tilt (polar angle χ, 0–88°) and rotation (azimuthal angle ϕ, 0–360°) of the sample with respect to the X-ray beam to perform texture goniometry measurements, where angular step widths of 1° in both χ and ϕ were adjusted. The rocking curve measurement (ϕ-scan) for the determination of the crystallite twist component was done with a high-resolution diffractometer equipped with an in-plane diffraction arm for in-plane measurements using a parallel beam. The in-plane ϕ-scans were performed at grazing incident beam of 0.25° to the sample surface (horizontal).

For cathodoluminescence (CL) measurements, a scanning electron microscope equipped with a helium cryostat (6 K) was applied. The microscope was operated at 8 kV acceleration voltage and a beam current of 1 nA. A monochromator with 150 g/mm grating and a nitrogen cooled 1024×256 pixels back illuminated UV enhanced CCD detector with a pixel size of $26\,\mu m \times 26\,\mu m$ was used.

TEM specimen preparation is essential for atomic-resolution Cs-corrected STEM imaging. Due to the different hardnesses of the substrate material 6H–SiC (22.9 GPa at 300 K) (Yonenaga, 2001) and the film material GaN (10.8 GPa at room temperature) (Yonenaga et al., 2000), the conventional preparation of TEM specimens is challenging. For STEM investigations, cross-sectional TEM specimens were prepared by focused ion beam (FIB) technique. The FIB lamellae were cut out using a Ga ion beam with beam energy of 30 keV and beam current of 4 nA. Afterwards, the lamellae were processed to electron transparency with a 5 keV Ga ion beam at a beam current of 50 pA. To reduce the TEM specimen thickness further and to improve the surface quality, the lamellae were polished with a focused argon ion beam in a NanoMill system (Model 1040, Fischione Instruments). Ion energies from 900 eV down to 400 eV were used to remove implanted Ga ions and amorphous regions caused by the FIB process (Poppitz et al., 2014). The FIB lamellae were oriented in $[2\bar{1}\bar{1}0]$ zone axis for hexagonal GaN (w-GaN) and 6H–SiC and in $[01\bar{1}]$ zone axis for cubic GaN (z-GaN).

STEM work was performed with a probe Cs-corrected TEM (Titan cubed G2 60-300) operated at 300 kV. A high-brightness electron gun (X-FEG), bright-field (BF), annular dark-field (ADF), high-angle annular dark field (HAADF) STEM detectors as well as a post-column Gatan imaging filter (GIF Quantum 963/P with DualEELS and fast shutter) and a SuperX detector for EDX analysis are available in the microscope. By working in STEM mode with an acceleration voltage of 300 kV, a point resolution of 70 pm can be achieved. For imaging of light elements at the GaN–SiC interface, annular BF (ABF) method was used. Measurements of TEM specimen thickness were performed by electron energy loss spectroscopy. Before STEM work, the TEM specimen was treated in a plasma cleaner for 10 min with a H_2/O_2 plasma recipe. All STEM images were slightly digitally filtered by a Wiener filter for noise reduction. Annular ranges of 79.5–200 mrad for the HAADF detector, 19–106 mrad for the ADF detector and of 10–19 mrad for