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Molecular Beam Epitaxy of Cubic Group III-Nitrides on free-standing 3C-SiC substrates

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Abstract. Cubic GaN, $Al_xGa_{1-x}N$/GaN and $In_yGa_{1-y}N$/GaN multiple quantum well (MQW) layers were grown by plasma assisted molecular beam epitaxy on 200 µm thick free standing 3C-SiC substrates. The influence of the surface roughness of the 3C-SiC substrates and the influence of metal coverage during growth are discussed. Optimum growth conditions of c-III nitrides exist, when a one monolayer Ga coverage is formed at the growing surface. The improvement of the structural properties of cubic III-nitride layers and multilayers grown on 3C-SiC substrates is demonstrated by 1 µm thick c-GaN layers with a minimum x-ray rocking curve width of 16 arcmin, and by c-AlGaN/GaN and c-InGaN/GaN MQWs which showed up to five satellite peaks in X-ray diffraction, respectively.

Introduction

The absence of polarization fields in cubic III-nitrides may be advantageous for device applications; however, the metastability of the cubic phase imposes stringent conditions on growth procedure [1]. Recently, high quality, bulk-like 3C-SiC substrates have been fabricated by HOYA Advanced Semiconductor Technologies Co., Ltd [2]. On such 3C-SiC substrates molecular beam epitaxy (MBE) has been performed with the goal to improve significantly the structural perfection of cubic III-nitrides and to enhance the interface quality of layered structures like superlattices (SLs), distributed Bragg reflectors (DBRs) and multi quantum wells (MQWs).

Experimental

Cubic group III-nitride samples were grown on 200 µm thick, free standing 3C-SiC(001) substrates by molecular beam epitaxy (MBE). An Oxford Applied Research HD25 radio frequency plasma source was used to provide activated nitrogen atoms. Indium, aluminium and gallium were evaporated from Knudsen cells. Prior to growth, the 3C-SiC substrates were chemically etched by organic solvents and a buffered oxide etch (BOE) and annealed for 10 hours at 500°C. Cubic GaN layers were deposited at 720°C directly on 3C-SiC substrates. The adsorption and desorption of metal (Ga) layers on the c-GaN surface was investigated using the intensity of a reflected high energy electron beam (RHEED) as a probe. Cubic $Al_xGa_{1-x}N$/GaN multiple quantum wells (MQWs) and cubic $In_yGa_{1-y}N$/GaN MQW were grown on such GaN buffer layers. The cubic $In_yGa_{1-y}N$/GaN MQW consists of 6 QWs with a barrier and well thickness of 12 nm and 5 nm, respectively. The $In_yGa_{1-y}N$ layers were deposited under In-rich conditions at a growth temperature of 610°C with an In content of 0.16.

The structural and morphological properties of both the 3C-SiC substrates and the group III-nitride epilayers were measured by high resolution X-ray diffraction (HRXRD) and atomic force microscopy (AFM). HRXRD and reciprocal space mapping (RSM) have been performed to determine the Al and In molar fraction and the strain in the $Al_xGa_{1-x}N$ and $In_yGa_{1-y}N$ layers.

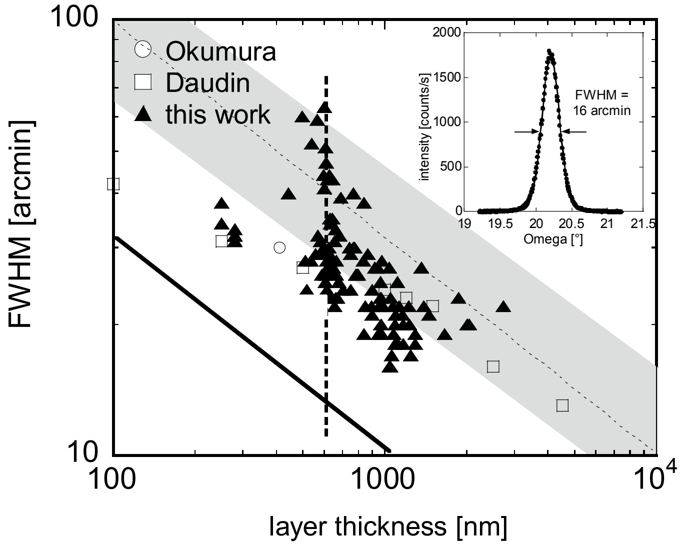

Fig. 1. Rocking curve linewidth of cubic GaN epilayers grown on 3C-SiC substrates versus thickness of the cubic epilayers. The inset shows a typical ω-scan of one of our cubic GaN epilayer. (triangles own data, circles data from Ref. [6], squares data from Ref. [7]).

Results and Discussion

The optimum conditions for the epitaxial growth of c-GaN are mainly determined by the surface morphology of the 3C-SiC substrate and the growth conditions during epitaxy, of which the surface stoichiometry and the substrate temperature are important parameters [3]. Since the growth parameters are interrelated, in-situ control of both substrate temperature and surface stoichiometry is necessary. This has been achieved by monitoring the MBE growth process by RHEED. Recently, optimum growth conditions of c-III nitrides were found, when a one monolayer Ga coverage is formed at the growing surface [4]. The RMS-roughness measured by a 5x5 μm^2 AFM-scan is decreasing from 16 nm to a minimum value of 2.5 nm and the width of the X-ray rocking curve of a 1 µm thick c-GaN layer grown with 1 monolayer coverage was about 16 arcmin (see inset of Fig. 1), which to our knowledge is one of the best values reported so far.

In Fig. 1 the rocking curve line width (ω-scan) of all our cubic GaN epilayers (full triangles) grown on 3C-SiC is plotted versus c-GaN layer thickness. Two clear effects can be seen from this plot. First a clear reduction of the full width at half maximum (FWHM) with increasing epilayer thickness is observed. This linewidth dependence is consistent with the defect annihilation process observed in cubic GaN grown on GaAs (001) substrates [5]. Since in zinc-blende structure the stacking faults (SFs) lie on the (111) planes, an annihilation mechanism is possible, when two SFs, lying, for example on the (111) and on the (-1-11) planes intersect and annihilate simultaneously with the creation of a sessile dislocation aligned along [110] directions. For the case of 3C-SiC, where the lattice mismatch is only –3.7 % to cubic GaN, the full line shows the theoretical calculated FWHM as a function of layer thickness using the dislocation glide model by Ayers [6]. This model implies that the dislocation density N_{disl} is inversely proportional to the layer thickness d and that the FWHM is proportional to $d^{-1/2}$. Comparing the full curve with the experimental data (full triangles) a reduction of the FWHM by a factor 1.5 is still possible. For the sake of completeness the shaded area in Fig. 1 depicts the range of FWHM values as measured for cubic GaN grown on GaAs substrates. By comparing our data with data cited in literature the dependence of the FWHM on film thickness has to be taken into account. Up to now, only two other groups reported data on 3 - 5 µm thick 3C-SiC/Si (001) pseudo-substrates grown by chemical vapor deposition (open circle Ref. [7] and open squares Ref. [8]). As can be seen in Fig.1 we are clearly able to remain under the best cited values, indicating the improved structural quality of our c-GaN epilayers due to the availability of free standing, bulk like 3C-SiC (001) substrates.

As mentioned above, the second parameter which strongly influences the quality of the cubic epilayers is the roughness of the 3C-SiC substrate. In Fig. 1 the rocking curve linewidth of 600 nm thick cubic GaN epilayers varies from 60 arcsec to 20 arcsec for 3C-SiC substrates with different surface roughness (indicated by the vertical dashed line). The root mean square (RMS) roughness of the corresponding 3C-SiC substrates as measured by AFM on 5 x 5 μm^2 large areas varied between 11 nm and 0.7 nm, respectively. The smoother the surface of the substrates the narrower is the FWHM and the higher is the structural quality of the cubic epilayer. This observation is in agreement with measurements of Ref. [7] and indicates that a further improvement of the surface preparation (atomic flat surfaces) may allow to reach the theoretical limit indicated by the full line.

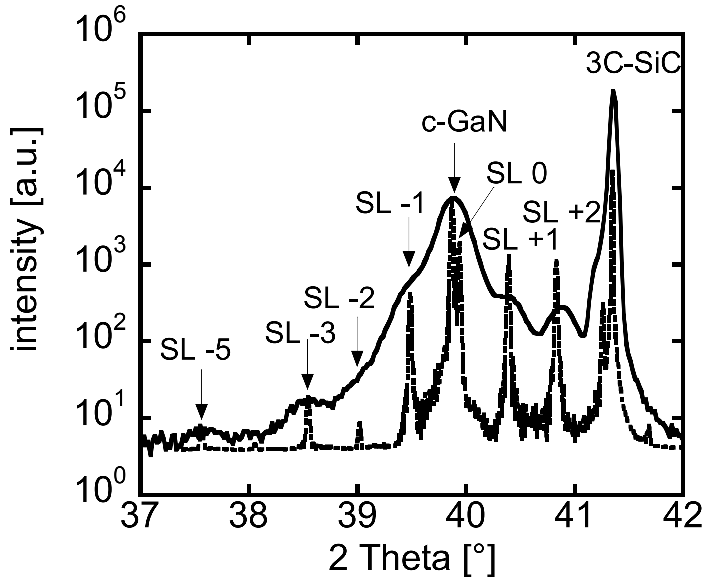

Fig. 2. Measured ω–2Θ scan of 15 x $Al_{0.3}Ga_{0.7}N$/GaN MQW structure (solid line) and simulated data (dotted line). The well and the barrier width are 10.2 nm and 10.8 nm, respectively.

On such optimized GaN epilayers cubic $Al_xGa_{1-x}N$ epilayers and cubic $Al_xGa_{1-x}N$/GaN MQW have been grown at 720°C. Such Al containing epilayers are necessary for cladding layers in laser diodes or LEDs and are also used in resonant cavity LEDs for the realization of $Al_xGa_{1-x}N$/GaN distributed Bragg reflectors.

Figure 2 shows the XRD ω-2Θ scan of the (002) Bragg-reflection of a 15 period $Al_xGa_{1-x}N$/GaN MQW structure (upper curve). The reflections of the 3C-SiC substrate, of the c-GaN buffer as well as several superlattice peaks (SL-5 to SL+2) are clearly seen. This indicates a good $Al_xGa_{1-x}N$/GaN interface quality. The experimental data have been fitted using dynamic scattering theory (lower curve), yielding a well width of 10.2 nm, a barrier width of 10.8 nm and an Al mole fraction of x=0.3. These values are in excellent agreement with data, which were obtained from growth rate measurements using RHEED oscillation period. The appearance of RHEED oscillations after opening the Al shutter [4] emphasizes a two dimensional $Al_xGa_{1-x}N$ growth mode at substrate temperatures of 720°C.

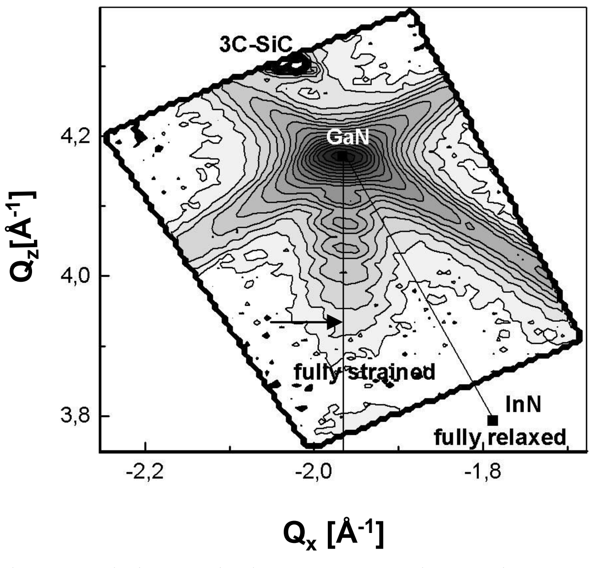

Fig. 3. High resolution XRD reciprocal space map near (-1-13) reflection of a 6 fold cubic $In_{0.16}Ga_{0.84}N$/GaN MQW grown on free standing 3C-SiC substrates. Arrow indicates 5th order superlattice peak.

For optoelectronic devices like high efficient green or blue LEDs $In_yGa_{1-y}N$/GaN quantum wells are the key elements in the active zones. In hexagonal $In_yGa_{1-y}N$/GaN MQWs, which are mainly grown in a (0001) growth direction, strong built-in electric fields due to the piezoelectric effect and spontaneous polarization severely reduce the radiative recombination efficiency for wider QWs [9]. Due to the higher crystal symmetry polarization fields are absent in cubic III-nitrides and due to the slightly smaller energy gap of the cubic nitrides (200 meV lower than the hexagonal counterpart), smaller mole fractions of In in the well of $In_yGa_{1-y}N$/GaN quantum wells are necessary to reach emission wavelengths beyond 510 nm. Therefore, we have grown a 6-fold c-$In_yGa_{1-y}N$/GaN MQW on 3C-SiC substrates. Figure 3 shows a typical XRD reciprocal space map around the (-1-13) reflection on c-$In_yGa_{1-y}N$/GaN MQW. The superlattice peaks are clearly resolved up to the 5th order and the $In_yGa_{1-y}N$ wells are pseudo-morphically grown on the GaN barrier layers. From the X-ray diffraction data an $In_yGa_{1-y}N$ well thickness of 5.6 nm and a GaN barrier thickness of 12.1 nm are obtained. The In mole fraction is about 0.16. As recently reported such cubic $In_yGa_{1-y}N$/GaN MQWs show a strong and dominant room temperature emission at 2.4 eV (510 nm) [10]. These properties indicate that cubic $In_yGa_{1-}N$/GaN MQWs seem to be the material of choice for the realization of resonant cavity light emitting diode (RC-LED) for the 500–570 nm spectral range [11].

Summary

Cubic GaN epilayers and cubic $Al_xGa_{1-x}N$/GaN and $In_yGa_{1-y}N$/GaN MQWs were grown by plasma assisted molecular beam epitaxy on 200 µm thick free standing 3C-SiC substrates. The influence of the surface roughness of the 3C-SiC substrates and the influence of metal coverage during growth are studied in detail. Optimum growth conditions of c-III nitrides were found, when a one monolayer Ga coverage is formed at the growing surface. X-Ray diffraction revealed superlattice peaks up to the 5th order indicating smooth interfaces. The improvement of the structural properties of cubic III-nitride layers and multilayers grown on 3C-SiC substrates is demonstrated by a 1 µm thick c-GaN layer with a minimum x-ray rocking curve width of 16 arcmin, and by c-AlGaN/GaN and c-InGaN/GaN MQWs which showed up to five satellite peaks in X-ray diffraction. These results demonstrate that due to the availability of free standing, bulk like 3C-SiC (001) substrates and the improved structural quality of our cubic group III-nitride epilayers the way for future realization of optoelectronic and electronic devices is also opened for cubic nitrides.

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