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Growth of ternary and quaternary cubic III-nitrides on 3C-SiC substrates

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Cubic GaN, $Al_xGa_{1-x}N$/GaN multiple quantum wells and quaternary $Al_xGa_yIn_{1-x-y}N$ layers were grown by plasma assisted molecular beam epitaxy on 3C-SiC substrates. Using the intensity of a reflected high energy electron beam as a probe optimum growth conditions of c-III nitrides were found, when a 1 monolayer Ga coverage is formed at the growing surface. Clear RHEED oscillations during the initial growth of $Al_xGa_{1-x}N$/GaN quantum wells were observed. X-ray diffraction measurements of these quantum well structures show clear satellite peaks indicating smooth interfaces. Growth of quaternary $Al_xGa_yIn_{1-x-y}N$ lattice matched to GaN were demonstrated.

1 Introduction

It is well established that group III-nitrides with cubic crystal structure can be grown on substrates with cubic structure [1]. The absence of polarization fields in c-III nitrides may be advantageous for some device applications, however, the metastability of the cubic phase imposes stringent conditions on the growth procedure. The molecular beam epitaxy (MBE) of c-III nitrides has been studied extensively, mainly using GaAs as substrates, which, due to their low thermal stability and the large lattice misfit, have limited the structural perfection of the epitaxial layers [2]. However 2-inch 3C-SiC substrates became available allowing substantial improve of the quality of the c-III nitride structures.

In this paper we present results of the MBE of cubic GaN and $Al_xGa_{1-x}N$/GaN MQWs on 3C-SiC substrates. Growth was carried out under well defined Ga-rich condition revealing a pronounced reduction of surface and interface roughness. We also report the first growth of quaternary c-III nitrides, which are lattice matched to cubic GaN.

2 Experimental

All the samples were grown on 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. 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) were grown on 650 nm thick GaN buffer layers. Quaternary $Al_xGa_yIn_{1-x-y}N$ layers were grown on a 500 nm c-GaN buffer at 620 °C.

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Structural characterization was carried out by High Resolution X-Ray Diffraction (HRXRD) and Atomic Force Microscopy (AFM). The composition of quaternary $Al_xGa_yIn_{1-x-y}N$ alloys was evaluated using Energy Dispersive X-Ray Analysis (EDX).

3 Results and discussion

The optimum conditions for the epitaxial growth of c-GaN are mainly determined by two parameters, the surface stoichiometry and the substrate temperature [1]. Both parameters are interrelated; therefore an in-situ control of both substrate temperature and surface stoichiometry is highly desirable. This has been achieved by monitoring the MBE growth process by RHEED. The study of the surface reconstruction behaviour was one of the key issues in understanding the c-III nitride growth [2–4]. First principle calculations by Neugebauer et al. [3] show that all energetically favoured surface modifications of the polar (001) c-GaN surface are Ga-stabilized and therefore optimum growth conditions are expected under slightly Ga-rich conditions.

We have obtained the Ga coverage of c-GaN by measuring the intensity of the reflected electron beam ((0,0)-streak RHEED intensity). During growth interruption the adsorption and desorption of gallium on a c-GaN (001) surface was measured employing the intensity of a reflected high energy electron beam (RHEED intensity) as a probe. After opening the Ga shutter we observed a decrease of the RHEED intensity to a saturation value, which was related to the impinging Ga flux. Using the known value of the Ga-flux and the transient time of the RHEED intensity we were able to estimate the amount of adsorbed gallium. After closing the Ga shutter the RHEED intensity reached the starting value indicating a total desorption of excess Ga.

A similar procedure was used to measure the Ga coverage during c-GaN growth. Figure 1 shows the RHEED intensity measured after opening the N shutter with the Ga flux on. From the drop of the RHEED intensity after the nitrogen shutter was opened we were able to measure the Ga-coverage during growth with an accuracy of 0.1 monolayer (ML). Details of this procedure will be described elsewhere. The influence of the Ga-coverage during growth on the roughness of c-GaN layers is depicted in Fig. 2. We find that the RMS-roughness measured by a 5x5 μm^2 AFM-scan is decreasing to a minimum value of 2.5 nm with increasing Ga-coverage (up to 1 monolayer). The width of the (002) X-ray rocking curve

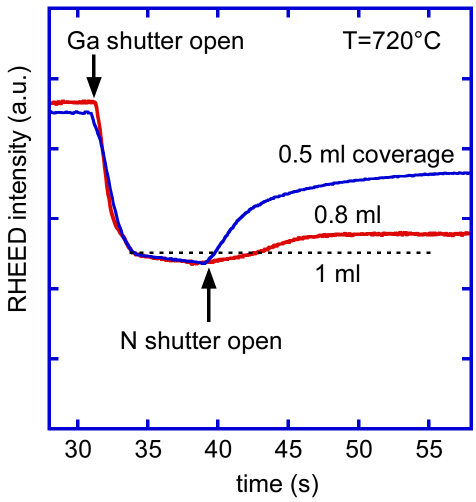

Fig. 1 Measured RHEED intensity during the initial growth of c-GaN. The RHEED intensity after opening the N shutter yields the amount of excess Ga on the c-GaN surface.

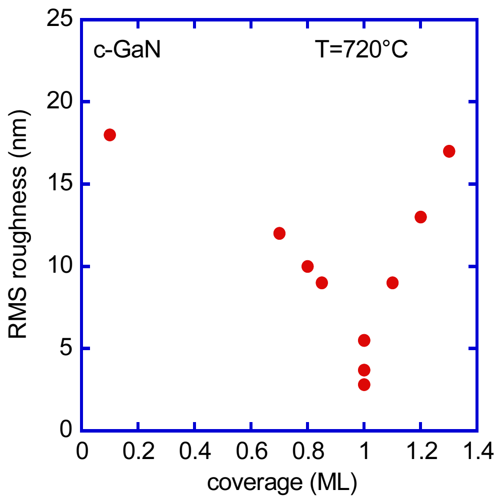

Fig. 2 Measured RMS roughness of c-GaN layers versus the Ga coverage. Minimum roughness is achieved at a coverage of 1 ML.

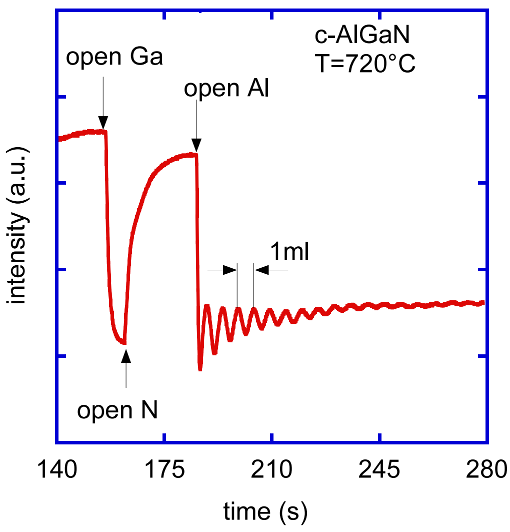

Fig. 3 Measured RHEED intensity during the initial growth of $Al_xGa_{1-x}N$. The RHEED oscillations during $Al_xGa_{1-x}N$ growth reveal a growth rate of 158 nm/h.

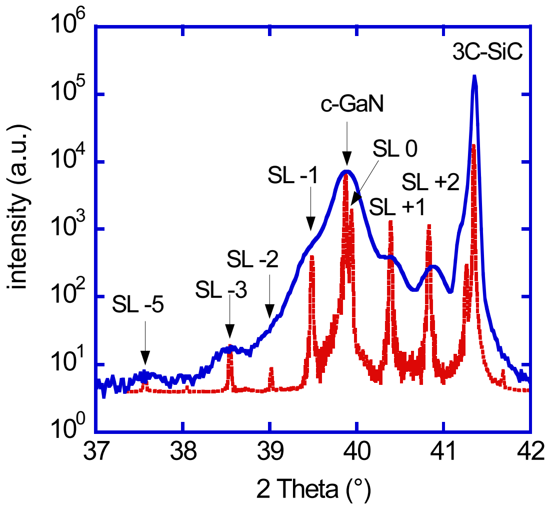

Fig. 4 Measured ω–2Θ–scan of a 15-fold $Al_{0.3}Ga_{0.7}N$/GaN MQW structure (solid line) and simulated data (dotted line). The well and the barrier width are 10.2 and 10.8 nm, respectively.

measured in double axis configuration of a 1 µm thick c-GaN layer grown with 1 monolayer coverage was about 16 arcmin. The FWHM of c-GaN layers is decreasing with increasing thickness indicating a decrease of the dislocation density due to annihilation of dislocations in cubic GaN [1]. Among those c-GaN layers with equal thickness of 1 µm 16 arcmin is a best value. A further increase of the Ga-coverage leads to a pronounced increase of the roughness, which may be due to accumulation of Ga on the surface.

The RHEED intensity was also monitored during the initial growth of $Al_xGa_{1-x}N$/GaN quantum well structures. An example is shown in Fig. 3. After opening the Al shutter clear RHEED oscillations were observed, indicating a two dimensional $Al_xGa_{1-x}N$ growth mode at substrate temperatures of 720 °C. The surface diffusion length of aluminium is smaller than that of gallium, therefore one would expect RHEED oscillations with the growth of GaN rather then with AlGaN. However, our experiments show the opposite behavior. The reason for that is not clear, it may be due to a kind of “surfactant” effect of Ga on the (001) surface, similar to what has been reported for In on hexagonal GaN [5].

Figure 4 shows the XRD ω–2Θ-scan of the (002) Bragg-reflex of a 15 period $Al_xGa_{1-x}N$/GaN MQW structure. The reflexes of the 3C-SiC substrate, of the c-GaN buffer as well as several superlattice (SL) peaks are clearly resolved. We were able to resolve SL peaks up to the 5th order. The broadening of the peaks is mostly dominated by the interface roughness, which is in the order of 2-3 nm. The experimental data have been fitted using dynamic scattering theory, 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.

First quaternary alloys were grown at 620 °C. It is well known from c-$In_xGa_{1-x}N$ growth that only substrate temperatures below about 650 °C allow the incorporation of reasonable amounts of indium [6]. The EDX spectrum of an $Al_xGa_yIn_{1-x-y}N$ layer is plotted in Fig. 5. The peaks correspond to gallium $K_{\alpha1}$, aluminium $K_{\alpha1}$ and indium $L_{\alpha1}$ X-ray emission, respectively. From the measured intensities we get the AlN and GaN mole fractions, which are x = 0.12 and y = 0.84, respectively. The InN mole fraction is 0.04. With the lattice parameters of cubic nitrides [1] a_{AlN} = 4.38 Å, a_{GaN} = 4.52 Å and a_{InN} = 4.98 Å we can calculate the lattice parameter of the $Al_{0.12}Ga_{0.84}In_{0.04}N$ layer by Vegard’s law

$$a_{AlGaInN} = xa_{AlN} + ya_{GaN} + (1-x-y)a_{InN} \,. \qquad (1)$$

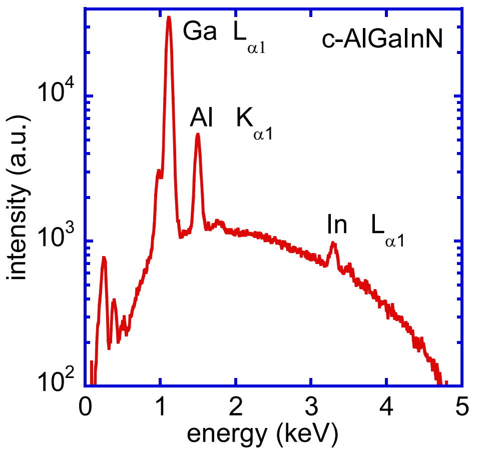

Fig. 5 EDX spectrum of a $Al_{0.12}Ga_{0.84}In_{0.04}N$/GaN heterostructure. The peaks correspond to the indicated X-ray emission.

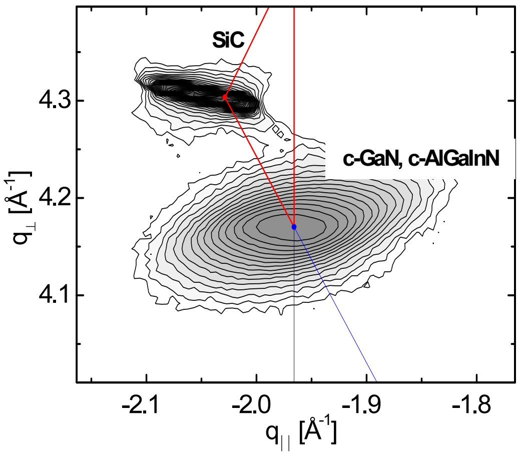

Fig. 6 Reciprocal space map of the (-1-13)-reflex of a $Al_{0.12}Ga_{0.84}In_{0.04}N$/GaN heterostructure. The Al-GaInN reflex is super positioned to the GaN reflex indicating lattice matching.

We find that the lattice constant $a_{AlGaInN}$ = 4.52 Å is identical to that of the GaN buffer. Figure 6 shows the reciprocal space map of the c-$Al_{0.12}Ga_{0.84}In_{0.04}N$ /GaN heterostructure. Two Bragg reflexes, one of the3C-SiC substrate and one which is due to the c-$Al_{0.12}Ga_{0.84}In_{0.04}N$ /GaN heterostructure are observed. The superposition of the c-$Al_{0.12}Ga_{0.84}In_{0.04}N$ and the GaN Bragg peak indicate clearly, that both layers have identical lattice parameters, in good agreement with the results of the EDX composition measurements. The RSM in Fig. 6 shows multiple peaks for the 3C-SiC substrate due to small angle grain bounderies. They may also cause some broadening of the GaN-AlGaInN reflex, however we are not able to resolve the reflexes from different grains.

4 Conclusion

The roughness of the surface and the interfaces of c-III nitride heterostructures grown by MBE on 3C-SiC substrates was significantly reduced by growth under controlled Ga-excess conditions. The amount of excess Ga was obtained from RHEED intensity measurements. During growth of $Al_xGa_{1-x}N$/GaN MQW clear RHEED oscillations were observed allowing a stringent control of the growth rate. X-ray diffraction revealed superlattice peaks up to the 5th order indicating smooth interfaces. First growth of quaternary $Al_{0.12}Ga_{0.84}In_{0.04}N$ lattice matched to GaN is demonstrated.

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