In-Situ Growth Monitoring and On-Line Composition Determination of MOCVD GaN by Spectroscopic Ellipsometry

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In-situ ellipsometry measurements during MOCVD growth of GaN and its ternary compounds AlGaN and InGaN were performed. We show that individual process steps can be identified and resolved with much higher detail than with currently available reflectometry setups. Using the Virtual Interface Approximation (VIA), we are also able to calculate the Al content of the growing layer in real time with good accuracy. For In containing samples we observe a rather large drift originating from a concentration gradient in the layer due to the immiscibility in the GaN/InN system.

Introduction

Group III nitrides have attracted tremendous R&D effort in the past few years resulting in the commercialization of optoelectronic devices operating in the blue and ultraviolet spectral range. Other fields of application include high mobility transistor devices (HEMT's), UV detectors and laser diodes for tomorrow's range of optical storage products [1]. The fabrication of GaN layers is mostly done by Metalorganic Chemical Vapor Deposition (MOCVD), which provides high growth rates and throughput in a non-UHV environment, both of which are well suited for industrial size production. The range of real-time diagnostic tools for MOCVD reactors is however quite limited, as electron diffraction techniques like RHEED (reflection high energy electron diffraction) cannot be used at atmospheric pressure. Optical methods like spectroscopic ellipsometry (SE) or reflection difference spectroscopy (RDS) have been successfully employed for in-situ monitoring of III-V and II-VI compounds. [2], [3]

We show the successful installation of a spectroscopic ellipsometer to a commercial MOCVD reactor and that these measurements have considerable advantages over reflectometry setups regarding sensitivity and an improved signal-to-noise ratio. Our further attention was focused towards the on-line monitoring of ternary alloy compositions in AlGaN and InGaN layers. The Virtual Interface Approximation (VIA) provides an easy and accurate way of determining the dielectric function of a growing layer which can be correlated to a certain Al content by cross-calibration with ex-situ methods like high resolution X-ray diffraction (HRXRD) or secondary ion mass spectroscopy (SIMS).

Experimental Setup

For the growth of group III nitrides, we use a modified commercial AIXTRON AIX200RF-S reactor with extra viewports to accommodate a spectroscopic ellipsometer. Strain free windows are necessary to avoid stress-induced polarization effects on incident and measured light beams. SE measurements were made with an ISA Jobin Yvon ellipsometer operating in the spectral range of 1.5 – 5.2 eV. Additional characterization was done with a laser reflectometer operating at 1.86 eV.

Figure 1 shows reflectivity and kinetic ellipsometry measurements during the growth of a hexagonal GaN layer on sapphire (0001) substrates. Both measurements have been performed below the fundamental absorption edge of GaN to avoid damping of thickness oscillations. We can clearly identify the important steps necessary to achieve high quality layers [2]:

(a) Heating of the substrate to 1200°C: the reflectometry signal increases due to thermal emission of the sample

(b) Desorption under H_2 atmosphere at 1200°C

(c) Decrease of temperature to 540°C for the deposition of low-temperature GaN nucleation layer

(d) Growth of nucleation layer: an islanded film covering the substrate surface is formed and an oscillation representing the first thickness fringe is recorded

(e) Annealing of nucleation layer through increase of the temperature to 1050°C: The behavior of both signals originates from the coalescence of GaN islands and a roughening of the surface. Spectroscopic measurements before and after annealing show layer thicknesses of 50nm and 30 nm respectively.

(f) Growth of GaN at 1050°C with low growth rate (large period oscillations in both reflectometry and SE)

(g) Deposition of GaN with a growth-rate of ~3 µm/h.

It can easily be seen that all the information contained in the reflectometry signal is also contained in the ellipsometry measurement. In this stage, ellipsometry offers the advantage of being independent of absolute intensities (only polarization and phase change are measured), which makes it immune to influences like wobbling of the sample and thermal emission at high temperatures.

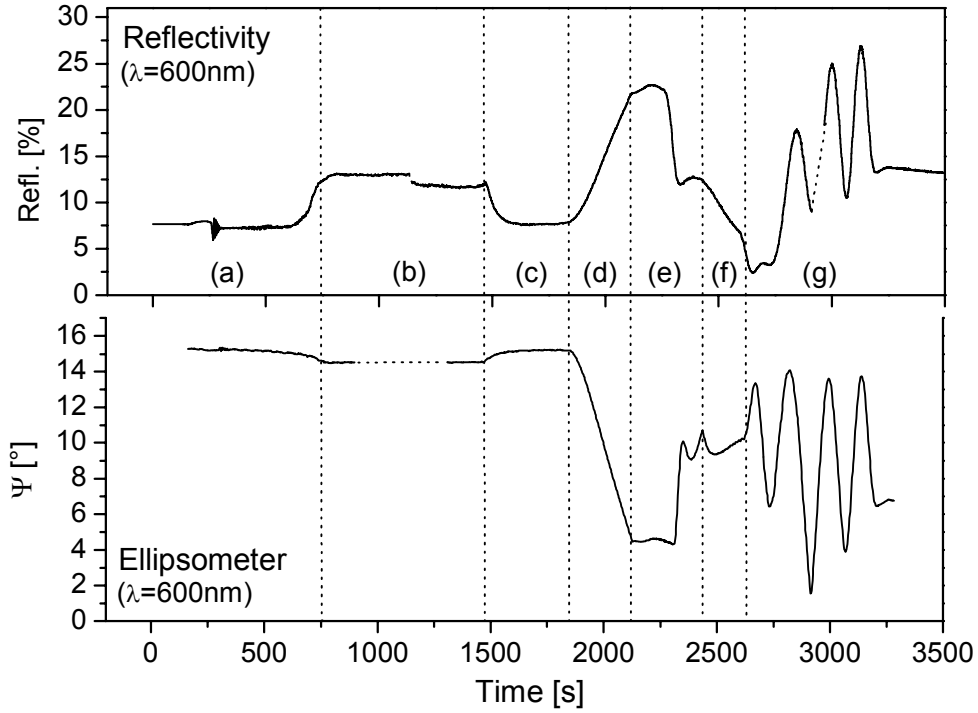

Fig. 1: Reflectometry vs. ellipsometry during the growth of hexagonal GaN layers on sapphire.

Concentration Monitoring

To relate optical information to sample parameters like ternary alloy compositions, it is necessary to perform complementary measurements and relate them to optical data. In our case, we performed high-resolution x-ray diffraction (HRXRD) measurements to gather data about the composition and stress and strain state of the samples. The precision in concentration from these measurements is in the range of ±0.5%.

As there is very little published data of optical constants of these compounds at growth temperature, we had to determine first the dielectric responses of layers with different Al content. Together with the exact concentration values from XRD, it was used as the input data for an algorithm based on the virtual interface approximation (VIA), which determines Al concentrations in real time [4]. The main idea of the VIA is to divide the sample structure into two parts, namely an overlayer and a so-called pseudosubstrate representing the whole structure below. From this 3-layer system (substrate, overlayer, ambient), the dielectric response of the overlayer can be calculated at each time step. The input parameters are the angle of incidence, the monitoring wavelength, the currently measured dielectric function, and its derivative with respect to the thickness of the growing layer. The last point implies the precise knowledge of the growth rate in the case of a measurement with a single wavelength only, which is accomplished by reflectometry in our case.

Figure 2 shows a typical kinetic measurement during the growth of an AlGaN layer. The dielectric function plotted in the complex plane describes an exponential spiral originating from the point for GaN and converging in the point for AlGaN. The inset shows an ellipsometric spectrum at growth temperature.

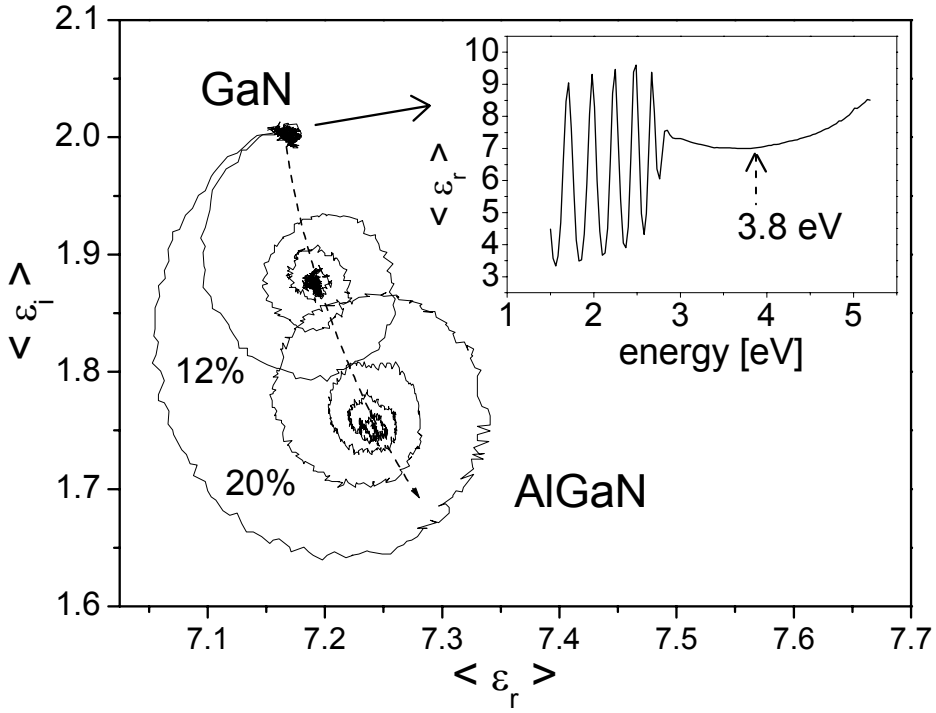

Fig. 2: Evolution of the pseudodielectric function during the growth of AlGaN layers with different Al contents on a GaN buffer. The inset shows a spectroscopic measurement at growth temperature (1050 °C).

A possible application of this method is the monitoring of superlattices as used in devices like vertical emitting laser diodes or transistors. Figure 3 shows the results of a

real-time measurement during the growth of a 30nm/50nm GaN/AlGaN superlattice with 12% Al in the barriers. Also shown in Fig. 3 is a composition profile of a structure with continuously varying Al content.

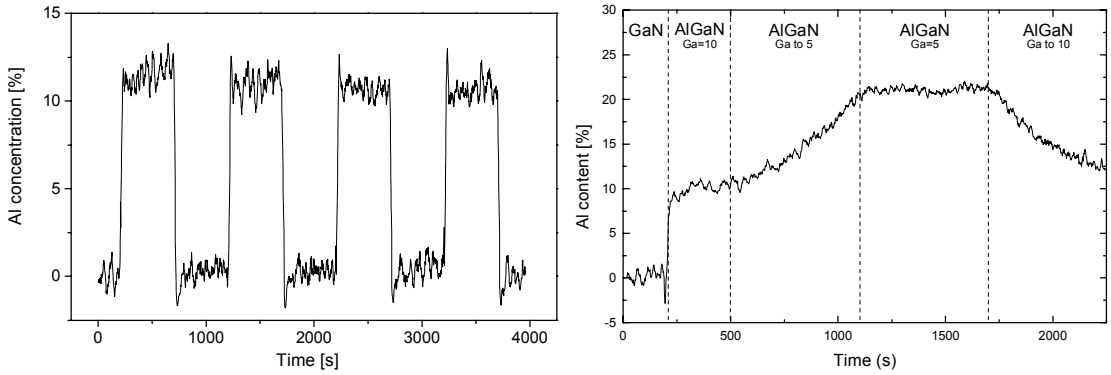

Fig. 3: Left: VIA calculated concentration in a GaN/AlGaN superlattice.
Right: Graded composition layer with Ga fluxes as indicated.

For In containing compounds, concentration monitoring is considerably more difficult. The system GaN – InN has a large miscibility gap resulting in a maximum concentration of a few percent in the crystal. Due to this behavior, InGaN layers also show rather large composition gradients in growth direction [5]. During in-situ measurements, the spirals of the dielectric function do not converge as nicely as for AlGaN compounds. Simulations show that this drift is due to a composition gradient in the layer.

Conclusion

We have demonstrated a method for the optical characterization of MOCVD growth processes of group III nitrides. Characterization of standard growth processes as well as accurate information about ternary layer compositions are obtained in real time thus yielding higher output and enhanced device quality.

Acknowledgements

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