In-situ X-Ray Diffraction during MOCVD of III-Nitrides: an Optimized Evaluation Algorithm

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Presently, we are able to measure in situ x-ray diffraction and spectroscopic ellipsometry simultaneously on rotating samples during the deposition process in our MOCVD reactor using a commercial available PANalytical Cu ceramic tube as x-ray source. Due to the natural wobbling of the rotating sample a compensation algorithm is used before adding up single spectra in order to improve signal to noise ratio before fitting procedure. In this paper we present an improved compensation algorithm based on a symmetric peak shape enabling the calculation of the peak symmetry axes from the centre of weight.

Introduction

Gallium nitride (GaN) and its alloys promise to be key materials for future heterojunction semiconductors devices aimed at high frequency, high power electronic applications [1]. However, manufacturing of such high performance products is challenged by reproducibility and material quality constraints that are notably higher than those required for optoelectronic applications. To meet this challenge, we already implemented, to our knowledge the first time, in situ x-ray diffraction (IXRD) for the metalorganic chemical vapor deposition (MOCVD) of III-nitrides as a real-time process tool using a standard x-ray source [2].

Presently, we are able to measure IXRD on rotating samples during the deposition process and in order to improve the signal to noise ratio a summation of single spectra is done. Because of the natural wobbling effects in MOCVD, all the single spectra are shifted with respect to each other. Therefore, a correction has to be done before adding up the channel intensities.

The algorithm used so far is based on peak maxima detection and the maximum intensity is interpreted as the peak centre defining the relative correction shift. In this paper we present a novel algorithm which takes into account also the surrounding of the peak maxima and results in more precise information on growth rate, composition and crystal quality.

Experimental Procedure

GaN and AlGaN layers are grown on c-plane sapphire substrates in an AIXTRON AIX 200~RF-S horizontal flow MOCVD reactor using standard techniques [3]. The sub-

strate is rotating in the gas flow (about 15 rotations per minute) in order to improve the homogeneity of the growing layer.

On the reactor shell two Be windows enable the incidence of the x-rays focused by a Johansson monochromator and the detection of the diffracted beam. The x-ray setup consists of a PANalytical Cu x-ray source and a commercially available multichannel detector of the type X'Celerator [4]. Furthermore, an in-situ multi-wavelength spectroscopic ellipsometer (SE) provides additional information during growth.

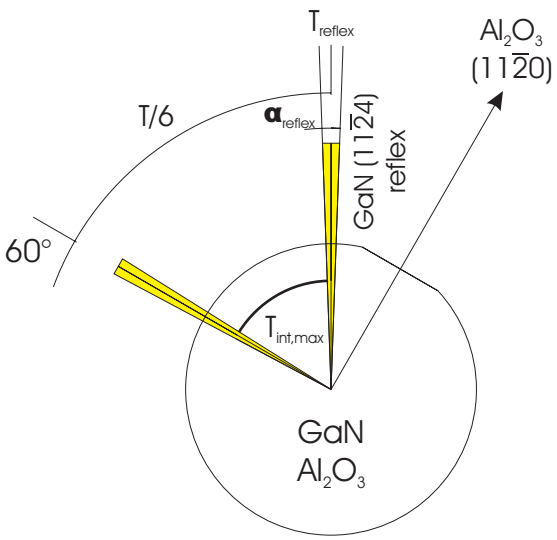

Fig. 1: Principal geometry of detected GaN (11-24) reflex (top view on the substrate).

The hexagonal GaN layer grows in the (0001) direction. The incident beam and the detector are adjusted in order to be sensitive to the (11-24) reflection of hexagonal GaN. Due to the hexagonal symmetry, the detector is illuminated six times per rotation period by a reflex which has an angular size defined by the angle α_{refl}. In connection with the rotation time of the sample (T) the time during which the reflex is visible for the detector (T_{refl}) is calculated by equation 1. To avoid a blur of the acquired spectrum only one GaN reflex must be included and this situation can be guaranteed by choosing a lower integration time than the critical integration time ($T_{int,max}$) which is defined by equation 1.

$$T_{int,max} = \frac{T}{6}\left(1 - T_{reflex}\right) = \frac{T}{6}\left(1 - \frac{\alpha_{refl}}{60}\right) \qquad (1)$$

On the one hand the use of integration times (T_{int}) below the critical time $T_{int,\ max}$ ensures that never more than one reflex is included in the resulting spectrum. On the other hand the reduction of the integration time leads to an increase of the probability (p_{empty}) to get empty spectra, decreasing in this way the efficiency of the system. Equation (2) defines p_{empty} as being proportional to T_{reflex} and the chosen integration time T_{int}.

$$p_{empty} = 1 - \left(T_{reflex} + T_{int}\right)\frac{6}{T} \qquad (2)$$

For the growth experiments, a constant integration time of about 90% of $T_{int,max}$ was used in order to ensure that only one reflex is included in the acquired spectrum, even if an increase of the rotation speed due to pressure or temperature changes in the growth chamber would take place.

In order to increase the signal to noise ratio, the single spectra have to be added up because of the unavoidable wobbling of the sample and due to the rotation during growth the spectra are shifted by an unknown angle in respect to each other. Previous works [2] report on an algorithm based on finding the maximum peak position (Pos_M) interpreted as the peak center and the point of reference. The results could be reasonable fitted at high enough spectral intensity expected at large layer thickness. An improved algorithm, working also at lower intensities, would represent a direct step toward an improved thickness resolution of the IXRD system.

For the results presented in this paper, we made use of a novel algorithm based on the fact that the reference peak has a symmetric shape. With this assumption the symmetry centre (Pos_{cw}) can be calculated from equation (3) by using the centre of weight of the reference peak. The detected maximum intensity of the spectrum (Pos_M) is taken as a first estimation of the centre of weight. The region around the maximum which should be included in the calculation can be defined by a constant (dk).

$$Pos_{cw} = \left(\sum_{k=Pos_M-dk}^{Pos_M+dk} I_k . x \right) \Bigg/ \left(\sum_{k=Pos_M-dk}^{Pos_M+dk} I_k \right) \qquad (3)$$

After compensation, the spectra are summed up and fitted by standard procedures [2].

Results and Discussion

By using the data acquired during a standard AlGaN growth process typical spectra are analyzed by both rotation compensating algorithms. After growing a GaN buffer on Al_2O_3 an $Al_{0.21}Ga_{0.79}N$ layer of about 500 nm is deposited and an IXRD spectrum is taken every 1.6 s. In order to improve the fitting results, 20 single spectra have been added after wobbling compensation and analyzed with both algorithms.

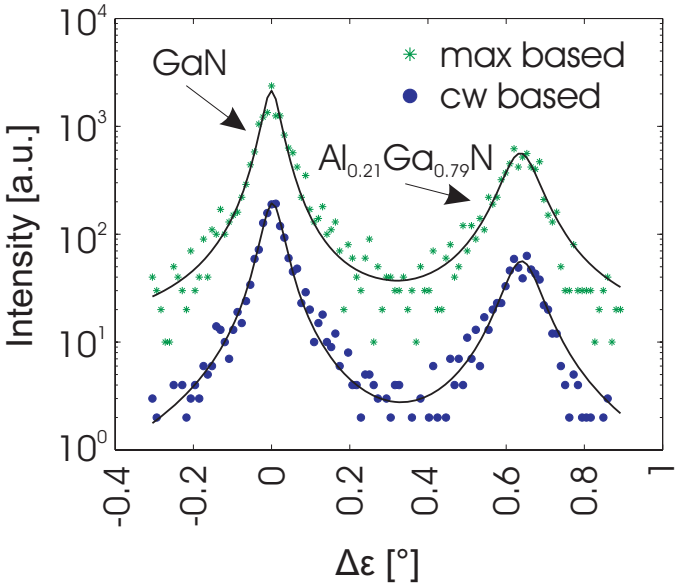

Fig. 2: 20 single spectra added up and fitted using the maximum based algorithm (star) and center of weight based algorithm (circles) as a function of angle Δε.

Figure 2 shows the intensity as a function of the diffraction angle of the scattered beam Δε relative to the GaN reference peak. A fit of the experimental points has been carried out by employing Pseudovoigt functions (solid lines).

For the top spectrum which is shifted for clarity by one order of magnitude represents the result by using the maximum based compensation algorithm. The fact that the channel with the maximum intensity is always shifted to $\Delta\varepsilon = 0$ results in a clearly visible overestimated peak intensity at this point. The lower spectrum (full dots) is the result of using the novel center of weight based algorithm which is not dominated by the previous described anomalous peak shape.

Summary

During MOCVD growth *in-situ* XRD spectra are acquired on rotating samples. In order to improve the signal to noise ratio the single spectra have to be added up before fitting procedure. Due to natural wobbling of the samples all single spectra are shifted in respect to each other which has to be compensated by an algorithm to avoid a blur of the added up spectra. We presented a new rotation compensation algorithm which is based on the symmetry of the peak all spectra are relatively shifted to. As a consequence the novel algorithm, taking into account also the surrounding of the peak maxima, results in a more precise information on growth rate, composition and crystal quality. Furthermore, the algorithm is more sensitive to small peak intensities, yielding an improved thickness resolution. The actual composition of the AlGaN layers could be determined already on a thickness of about 20 nm.

The obtained accuracy and the improved performance of the in-situ XRD setup represent a substantial step forward in the perspective of an effective closed-loop control of the MOCVD growth process.

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