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Effect of antimony on growth mode and properties of thick InGaN layers

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ABSTRACT

A comprehensive study on the effect of antimony on growth mode and crystal properties of thick InGaN layers grown by metal-organic vapor phase epitaxy is presented. Two growth regimes are identified by atomic force microscopy: while already minor antimony precursor flux induces the stabilization of $\{10\bar{1}1\}$ facets, the application of increased fluxes leads to the growth of nanoscopic islands which suppresses the typically observed V-pit formation and thus decreases the layer roughness. Further on, high incorporation rates of antimony cause to concentrations of up to 0.74%, at standard InGaN growth conditions, are revealed. The obtained results on the impact of antimony incorporation provide evidence that the generally assumed true surfactant behavior of antimony for InGaN growth has to be reconsidered since it significantly affects structural and luminescence properties.

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1. Introduction

Supplying a surface-acting agent (surfactant) during growth processes is a common option to influence the growth mode and optimize the structural properties of epitaxial layers [1]. The surfactant is supposed to alter the surface energy in order to facilitate the growth of distinct, smooth surfaces without being incorporated. In the case of III-nitrides, antimony has recently been proposed and investigated as a suitable surfactant to improve the homogeneity of (In)GaN layers [2–7], which are commonly used for opto-electronic devices such as light emitting diods (LED). The epitaxial growth of the InGaN-based quantum wells (QW) by metal-organic vapor phase epitaxy (MOVPE) is particularly challenging because of the pronounced miscibility gap between InN and GaN causing phase separation. The resulting indium content inhomogeneity leads to microscopic fluctuations of the band gap energy which hinder carrier diffusion, cause a broadening of the macroscopic emission wavelength and affect the efficiency of the LED [8].

According to recent reports, the addition of antimony during the growth of the QW can improve the optical quality [3,4] and might decrease the band gap energy [2]. However, published data concerning InGaN rely on the analysis of thin layers which exhibit a thickness of only several nanometers and are sandwiched between GaN layers. This complicates the analysis of the structural properties of the InGaN layer. For example, the extraction of the indium content by X-ray diffraction (XRD) is much more challenging than for thick layers [9]. Further on, the growth of thicker layers is required to enable the detectable formation of new growth facets in case of changed surface energies.

Addressing these topics, this work reports on obtained results concerning the growth of thick InGaN layers at different antimony fluxes. First, growth morphology and developing crystal facets are studied based on detailed atomic force microscopy (AFM) measurements. Two growth regimes are identified depending on the applied antimony flux. In the second step, the impact of antimony on indium incorporation and strain state of the InGaN layer is presented and discussed in terms of growth kinetics. Although antimony is generally assumed to be a surfactant in case of InGaN growth, results of compositional analysis provide evidence of significant antimony incorporation. A major impact of the antimony content on the emission wavelength as well as the crystalline properties is demonstrated.

2. Experimental

2.1. Growth details

All InGaN layers discussed in this work were grown by MOVPE on *n*-type GaN templates deposited on 4 in. c-plane sapphire substrates.

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The applied GaN buffer layers exhibited a thickness of 5 µm and a typical defect density of 5×10^8 cm^{-2}. Trimethylindium (TMIn), triethylgallium (TEGa), triethylantimony (TESb) and ammonia were used as precursors which were introduced in the reactor along with nitrogen as carrier gas. Thick InGaN layers were grown at 800 °C with a nominal thickness of 100 nm. Two sets of samples were investigated (Table 1). For the first one, the TESb/(TESb+TMIn+TEGa) (in short: TESb/III) molar flux ratio was varied (#1–#5) to evaluate the influence of an increasing TESb flux on InGaN growth mode and crystalline properties. The TMIn/(TMIn+TEGa+TESb) (in short: TMIn/III) molar flux ratio was kept constant at 22%. Two additional samples (#4a, #4b) were grown based on the sample with a TESb/III molar flux ratio of 0.133% (#4) to investigate possible effects of antimony on indium incorporation and to study the surfactant behavior of antimony with increasing TMIn/III molar flux ratio. All InGaN layers were grown without a capping layer and cooled down using an ammonia and nitrogen ambient to prevent alterations of the surface morphology by the desorption of indium or nitrogen.

2.2. Characterization

The morphology of the samples was analyzed by 2×2 µm^2 AFM scans using a Veeco Dimension Icon. The obtained data were processed by the *step line correction* and *plane level* functions of the Gwyddion software [10]. In distinct cases, the *facet analysis* function was applied to statistically study the orientation of analyzed facets. Therefore, each scanned position of the AFM image is evaluated by fitting a plane based on the height level of surrounding data points. It should be noted that only planes exceeding a scanned area of 23.4×23.4 nm^2 can be resolved considering the lateral resolution of the AFM image (3.9 nm per pixel) and the selected range (three data points) for the local plane fit. Furthermore, the geometry of the applied AFM cantilever model (Bruker TESPA) limits the discussed method to planes exhibiting inclination angles to 65°. To ensure stable measurement conditions, possible deterioration of the cantilever geometry was monitored before and after the measurements by scanning a reference sample. Further characterization of indium content, strain state and crystalline quality of the InGaN layers was carried out by performing XRD $0006\omega/2\Theta$ scans and $10\bar{1}5$ reciprocal space mappings (RSM) with a PANalytical X'Pert PRO. Selected samples were analyzed by liquid helium temperature photoluminescence (LT-PL) using a helium–cadmium laser with an excitation wavelength of 325 nm and secondary ion mass spectroscopy (SIMS). The SIMS measurements were done by a commercial material characterization laboratory which provided calibrated reference samples. Selected elements were probed as impurities in an InGaN matrix.

3. Results

3.1. Growth morphology of InGaN

The AFM analysis of the InGaN surface morphology of the TESb/III molar flux ratio series revealed two different growth regimes. As shown in Fig. 1, the introduction of minor TESb flux (marked as regime I) caused an increasing root-mean-square (RMS) roughness of the resulting surface compared to the reference InGaN sample. The corresponding AFM images depict a change in the growth morphology: While the InGaN reference (sample #1) features a smooth surface that is only interrupted by typical V-pits (Fig. 2a), sample #2 (Fig. 2b) already demonstrates the formation of nanoscaled ridges. Although the typical growth of III-antimonide compounds is only possible at temperatures below 650 °C [11,1], these observations suggest a significant impact of antimony altering the InGaN growth morphology. It can be assumed that adsorbed antimony atoms possess a high binding energy on the (0001) InGaN facet, which has been predicted by theoretical calculations of Gokhale et al. [7,6]. According to their results, antimony exhibits an even higher binding energy (BE(Sb)=-4.96 eV) than gallium (BE(Ga)= −3.55 eV) on the (0001) GaN surface. Thus, even the lowest TESb flow is sufficient to stabilize a distinct antimony adatom concentration on the growth surface, which significantly affects the growth morphology (regime I in Fig. 1).

Table 1
Overview of samples grown for the TESb/III molar flux ratio series by variation of the TESb flux. Samples of the TMIn/III molar flux ratios series (#4a and #4b) are based on sample #4 and feature a constant TESb flux, but increasing TMIn flux.

Sample	#1	#2	#3	#4	#4a	#4b	#5
TESb/III (%)	0	0.027	0.066	0.133	0.100	0.070	0.266
TMIn/III (%)	22	22	22	22	40	60	22

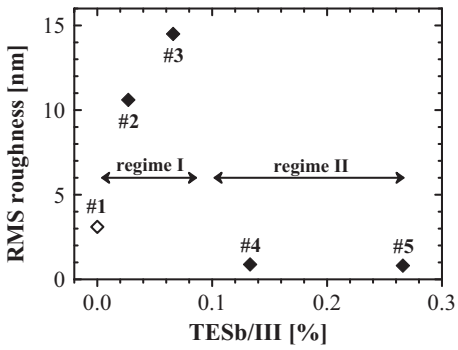

Fig. 1. Plot of obtained RMS roughness as a function of TESb/III molar flux (hollow symbol denotes reference InGaN layer).

Increasing the TESb/III molar flux ratio, the highest RMS roughness was obtained for sample #3. The corresponding AFM image (Fig. 2c) shows a transition from ridges towards the second growth regime, which is characterized by three-dimensional island growth. This island growth completely dominates the morphology of sample #4 (Fig. 2d). The size of the islands was significantly reduced, yielding a very smooth surface with an RMS roughness value of 0.9 nm (regime II in Fig. 1). Moreover, the formation of V-pits, which is typically observed during InGaN or GaN growth at the applied growth temperature of 800 °C [12,13], was completely suppressed. It can be concluded that a sufficient antimony adatom concentration was accumulated at the growth surface to induce a change in the surface reconstruction in regime II. This is in agreement with observations of Merrell et al. [2] for InGaN QW surfaces. In their study, AFM images indicated a comparable change of the growth mode with increasing TESb/III molar flux ratios although the InGaN layers featured only a thickness of 1.5 nm. Further on, the suggested existence of multiple surface reconstruction modes depending on the applied TESb flux has already been shown for GaP and InP [14,15]. However, to be able to detect a new dominant crystal facet, the growth of thick layers is required to facilitate the formation of the facet.

Investigating the crystallographic orientation of developing facets, the *facet analysis* function of the Gwyddion software was applied. Therefore, AFM data of selected samples of each growth regime were evaluated as described in Fig. 3a. The resulting histograms (Fig. 4) of the analysis are plotted in polar coordinates. Here, the radial coordinate corresponds to the inclination angle θ of the examined facet in comparison to the horizontal plane (zero tilt given by center of the plot), while the polar coordinate φ depicts the lateral tilt of a facet in reference to the flat orientation (all exemplary marked in Fig. 4b).

For the reference InGaN layer (Fig. 4a), the majority of identified facets show no tilt which is reasonable due to the dominating

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