Surfactants for the epitaxial growth of GaN (0001)

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The growth kinetics on epitaxial surfaces can often be significantly influenced by foreign atoms which are thermodynamically stable on the surface but are not incorporated into the growing film. Those species are called surfactants if the growth in terms of surface morphology or crystal perfection is improved. Theoretical investigations suggest indium as promising candidate for a surfactant on GaN [1], but only few experimental studies can be found in literature [2, 3, 4]. Surfactants for GaN are not only interesting to understand basic growth mechanisms of this material, but they may also help to overcome undesirable restrictions in growth conditions. For example, the growth of GaN by molecular beam epitaxy (MBE) has to be performed at lower temperatures compared to that by metalorganic vapour phase epitaxy (MOVPE) due to thermal decomposition of the material in vacuum. Low growth temperatures however impede step-flow growth which is prefered to obtain atomically smooth surfaces. Within this work different atom species as indium, bismuth and hydrogen are systematically investigated in view of their use as surfactant on GaN (0001) and the results are discussed with regard to theoretical predictions.

GaN layers were grown by MBE on thick GaN template layers deposited by MOVPE on *c*-plane sapphire. Indium and bismuth were evaporated by Knudsen cells whereas hydrogen was mixed to nitrogen and injected through a single rf plasma source. Care was taken to keep the active nitrogen flux constant when adding hydrogen by *in-situ* measuring the intensities of characteristic lines in the plasma discharge spectrum and by accurately monitoring the growth rate via ellipsometry. Epitaxial surfaces were investigated by reflection high energy electron diffraction (RHEED), X-ray photoelectron spectroscopy (XPS) and atomic force microscopy (AFM). Bulk properties of the grown films were studied using photoluminescence (PL) and transmission electron microscopy (TEM).

In a first step growth conditions were determined under which the different elements are stable on the GaN surface. Therefore intensity changes of the specular reflection in RHEED patterns under $[1\bar{1}00]$ incidence were recorded during deposition and free desorption of the elements as exemplarily shown in Fig. 1. During deposition of either gallium, indium or bismuth the signal drops which probably indicates a roughening of the

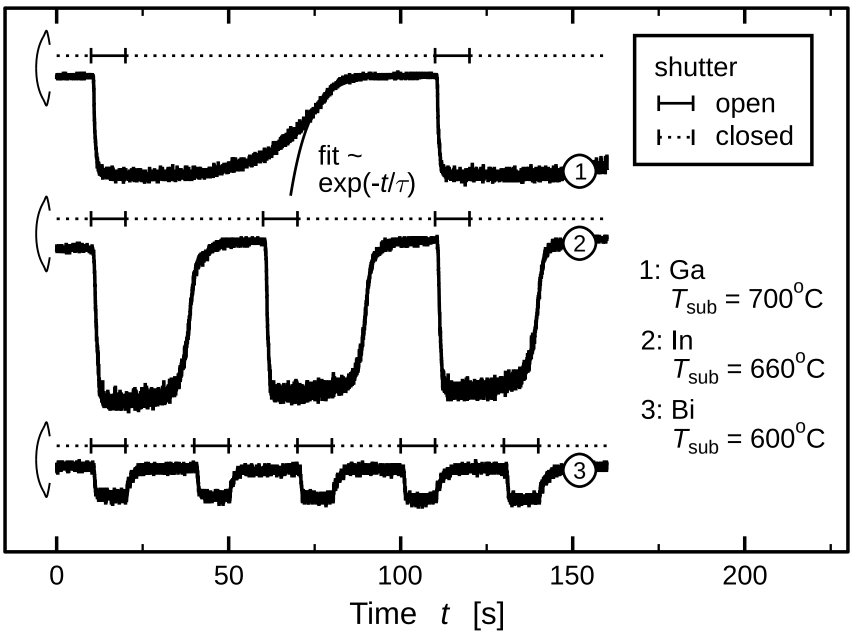

Fig. 1: Specular spot intensity in RHEED patterns with electron incidence along $[1\bar{1}00]$ during deposition and desorption cycles with gallium, indium and bismuth on GaN (0001). The corresponding shutter positions are indicated. An exponential fit is exemplarily shown for one transient.

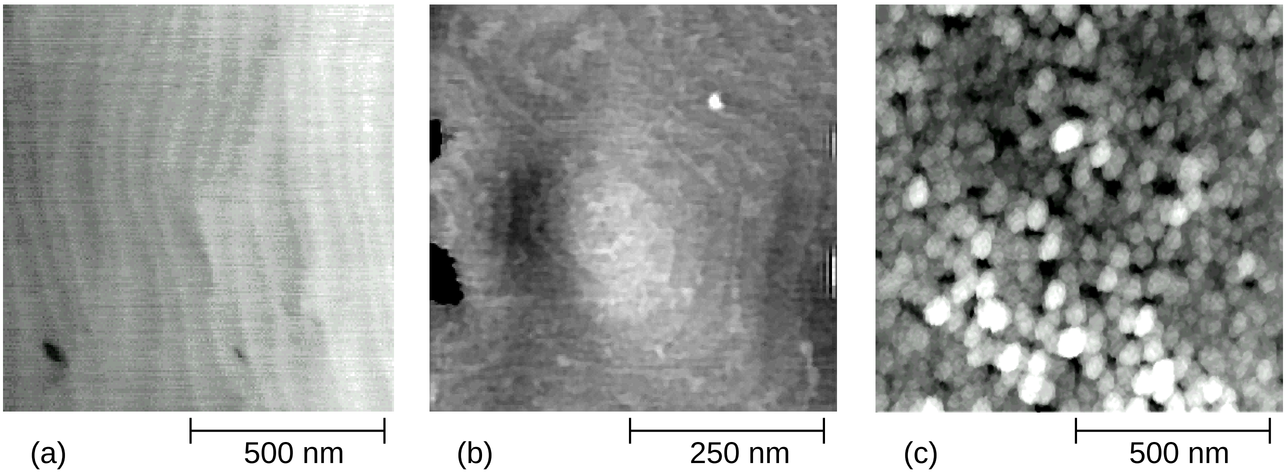

Fig. 2: AFM images of GaN surfaces: As-grown MOVPE template layer (a), 200 nm thick MBE layers grown under Ga rich conditions without (b) and with (c) the use of hydrogen.

surface. After stopping the deposition, excess material reevaporates and the surface recovers its original state. The final part of the slope before complete recoverage can be fitted to an exponential decay law providing a characteristic time constant for this process. The time constant is found to depend on the growth temperature as well as on the atom species. From Arrhenius plots an activation energy of 1.71 ± 0.05 eV could for instance be determined for the surface rearrangement after gallium deposition. This value is considerably smaller than the evaporation energy from liquid gallium, which is about 2.8 eV [5]. Even smaller values could be determined for indium and bismuth. They will be discussed in terms of material desorption from different states and the existence of diffusion barriers.

The impact of indium, bismuth and hydrogen on the growth kinetics and the defect formation in the film was studied by growing GaN films with and without a constant flux of the corresponding element and analyzing the samples *ex-situ*. A particular result for the use of hydrogen is depicted in Fig. 2, which shows surface images obtained by AFM. The starting surface before MBE growth is given by the MOPVE template shown in Fig. 2(a). It exhibits a well defined terrace structure with monolayer steps and pits at the terminating points of steps corresponding to threading dislocations. MBE layers deposited thereon at 660°C under Ga rich conditions grow in a twodimensional mode. Regarding to Fig. 2(b), spirals with hexagonal boundaries form on the surface, the steps of which again have monolayer height. With the addition of hydrogen, the surface becomes rougher and the terrace structure is lost as shown in Fig. 2(c). Instead grainy features have been formed which are rather typical for growth under nitrogen rich conditions [6]. However, accurate measurements of the growth rate confirmed that still the growth remained Ga-rich. Obviously, hydrogen serves as an anti-surfactant at this temperature, which impedes the surface diffusion of gallium or nitrogen or their incorporation at atomic steps. On the other hand, MOVPE growth in an hydrogen ambient is known to provide step-flow growth at significantly higher temperatures of 1000 to 1100°C. Therefore, samples similar to those presented in Fig. 2 were grown at different temperatures and their morphology is investigated. Moreover, the impact of indium and bismuth on growth performance is studied in the same way. PL and TEM measurements are underway to search for point or extended defects whose formation is either suppressed or supported by the use of the surfactant candidates.

[1] T. Zywietz, J. Neugebauer and M. Scheffler, Mat. Res. Soc. Fall Meeting, Boston (1999).

[2] F. Widmann, B. Daudin, A. G. Feuillet, N. Pelekanos and J. L. Rouviere, Appl. Phys. Lett. **73**, 2642 (1998).

[3] R. Klockenbrink, Y. Kim, M. S. H. Leung, C. Kisielowski, J. Kruger, G. S. Sudhir, M. Rubin and E. R. Weber, Mat. Res. Soc. Symp. Proc. **468**, 75 (1997).

[4] Y. Okamoto, S. Hashiguchi, Y. Okada and M. Kawabe, Jpn. J. Appl Phys. Lett. **38**, L230 (1999).

[5] R. E. Honig and D. A. Kramer, RCA Review **30**, 285 (1969).

[6] E. J. Tarsa, B. Heying, X. H. Wu, P. Fini, S. P. DenBaars and J. S. Speck, J. Appl. Phys. **82**, 5472 (1997).