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Structural investigation of growth and dissolution of $In_xGa_{1-x}N$ nano-islands grown by molecular beam epitaxy

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Abstract

Free-standing nanometre sized $In_xGa_{1-x}N$ islands and islands capped with thin GaN layers were grown by molecular beam epitaxy (MBE) and characterised by high resolution transmission electron microscopy (HRTEM). By adjusting the growth temperature of the $In_xGa_{1-x}N$ islands, deposition of the wurtzite modification was realised. The obtained free-standing islands are plastically relaxed by approximately equidistant misfit dislocations. For capped samples, the GaN cap layer thickness was varied between nominal thicknesses of 2 nm and 8 nm. Already for a 2 nm thick cap layer, strong dissolution of the islands occurred. In case of the 8 nm thick GaN cap layer, all former islands are dissolved, and an InGaN layer of approximately homogeneous indium concentration is observed.

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1. Introduction

Wurtzite InGaN quantum nano-structures have been employed as active material in light emitting diodes (LEDs) or laser diodes (LDs) [1–3]. In this context, the use of InGaN quantum dots (QDs) in the active region of e.g. LDs is especially promising due to the predicted improvement of the temperature stability or threshold current density of LDs. To obtain a high QD density as is required for QD-LDs, self-assembled growth of InGaN nano-islands is suitable. Possible approaches to obtain self-assembled InGaN islands are the utilisation of antisurfactants as e.g. Si [4], In [5], or air passivation [6], droplet epitaxy [7,8], or the reduction of the growth temperature for the InGaN islands in comparison to quantum wells (QWs). The latter approach, which was employed for this work, reduces firstly the diffusion length, thus three-dimensional growth is facilitated. Secondly, the dissociation and reevaporation of In is reduced. Consequently, more In can be incorporated in the growing material, which increases the stress and may lead to the Stranski–Krastanow growth mode [9], i.e. to a transition from a two-dimensional film growth to three-dimensional island growth.

For the application of QDs in high quality opto-electronic devices, the QDs need to be over-grown with a cap layer. The emitted wavelength of QDs depends not only on the composition, but also e.g. on their size and shape. All of these features can change during growth of a cap layer. Thus it is necessary to evaluate the corresponding changes of the composition and morphology of the InGaN nano-islands during over-growth. In this work, we use transmission electron microscopy (TEM) to study the formation of free-standing InGaN nano-islands and the change of the composition and geometry of the islands due to overgrowth with a GaN cap layer.

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Table 1
MBE growth parameters of the analysed InGaN island samples

Sample no.	T_g (InGaN) (°C)	t_g (InGaN) (s)	$\frac{In}{In+Ga}$ flux ratio	T_g (GaN, cap) (°C)	Nom. cap layer thickness (nm)
LT0	450	90	0.5	–	–
HT0	510	70	0.7	–	–
HT2	510	70	0.7	510	2
HT8	510	70	0.7	510	8

T_g denotes the growth temperature of the InGaN or the GaN cap layer and t_g is the duration of InGaN growth. Furthermore, the nominal GaN cap layer thickness is given.

2. Experimental details

All samples of this work were deposited on (0 0 0 1) sapphire substrates. On top, an about 2 μm thick GaN film was grown by metal organic vapour phase epitaxy (MOVPE). The subsequent growth was performed by molecular beam epitaxy (MBE) in an EPI 930 growth chamber. Prior to the deposition of InGaN, approximately 50 nm GaN were deposited.

The InGaN growth was in all cases under nitrogen rich conditions as this is known to promote three-dimensional growth [10,11]. The intended In concentration was 0.2. To obtain three-dimensional InGaN islands, a low growth temperature of about 500 °C was chosen. Detailed growth conditions are given in Table 1.

The growth of the InGaN layer was stopped in all cases after the reflection high energy electron diffraction (RHEED) pattern changed from streaky to spotty, indicating that a transition from two-dimensional film growth to three-dimensional InGaN island growth occurred. For the capped samples HT2 and HT8, the GaN cap layer was deposited under gallium rich conditions and at the same growth temperature as the InGaN to impede redistribution of In during the over-growth process.

The TEM specimens were prepared by a tripod method. Electron transparency was obtained by subsequent ion milling in a PIPS (Gatan) facility using Ar^+ ions at a voltage of 5 kV. For quantitative measurement of the In concentration, high resolution TEM (HRTEM) images were recorded at a CM20 UT transmission electron microscope equipped with a LaB_6 filament and operated at 200 kV. The In concentration was derived by strain state analysis of these images. Care was taken to ensure minimum electron beam exposure times of areas imaged for strain state analysis in order to minimise electron beam induced damage [12–14]. *Z*-contrast imaging was done at a Tecnai F20 ST equipped with a field emission gun (FEG) and a high angle annular dark field (HAADF) detector for scanning TEM (STEM).

3. Indium concentration measurement

For quantitative concentration measurement, strain state analysis [15] was performed, i.e. the In concentration was derived by combining Vegard's rule [16] and linear elasticity theory. Using the matrix notation, the strain $\vec{\varepsilon}$, the stress $\vec{\sigma}$ and the compliance $\mathbf{S}$ are connected via Hooke's law $\vec{\varepsilon} = \mathbf{S}\vec{\sigma}$. Assuming pseudomorphic growth, the shear components are neglected. For epitaxially grown islands without misfit dislocations, this approximation is only true for the bottom and the top of the InGaN islands, where finite element (FE) calculations show that the material is about completely strained to the a lattice parameter of the underlying GaN or completely relaxed, respectively. In between, a gradual relaxation of the island material takes place. Nevertheless, we found that the present islands are about completely relaxed due to misfit dislocations. Thus, neglecting shear is justified. Furthermore, taking into account that the component of the stress tensor in the [0 0 0 1] growth direction is zero, the actual tetragonally distorted lattice parameters $a^t(x)$ and $c^t(x)$ are connected via

$$c^t(x) = \left(1 + \varepsilon_3 \cdot \frac{a^t(x) - a(x)}{a(x)}\right) \cdot c(x) \tag{1}$$

with

$$\varepsilon_3 = \frac{C_{13}(r+1)(C_{12} - C_{11})}{C_{11}C_{33} - C_{13}^2 - rC_{12}C_{33} + rC_{13}^2}. \tag{2}$$

Here, $a(x)$ and $c(x)$ are the lattice parameters as derived from Vegard's rule, and r gives the degree of relaxation of a TEM specimen ($r = 1$: bulk limit for a thick specimen, $r = 0$: complete relaxed material in electron beam direction for a thin specimen). The components C_{ij} of the stiffness tensor $\mathbf{C} = \mathbf{S}^{-1}$ depend on the concentration and are supposed to vary linearly with x. For all calculations, the C_{ij} of the binary components are taken from Ref. [17], and the lattice parameters of GaN and InN are taken from Refs. [18,19], respectively. The used values are given in Table 2.

After inserting $C_{ij}(x)$ into Eq. (1), the In concentration x can be iteratively calculated if the tetragonally distorted lattice parameters are known. In case that $a^t(x)$ is known, for instance for pseudomorphic growth, the measurement of the tetragonally distorted $c^t(x)$ is sufficient to derive the local In concentration. This can be done by employing two beam imaging conditions in the TEM using the transmitted and the 0 0 0 2 beam and measuring the averaged projected displacements $\overline{u}$, i.e. averaged along the electron beam direction. These are normalised to a reference material of