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Growth and properties of InGaN nanoscale islands on GaN

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Abstract

Strong photoluminescence and radiative recombination lifetimes longer than 1 ns at room temperature have been observed in GaN/Si/InGaN/GaN structures containing InGaN submicron islands. The flat islands, with a width at their base in the order of 200 nm and a height in the order of 1–2 nm, grow in a spiral mode around dislocations with partial or pure screw character after a passivation of the GaN surface by a preflow of disilane. Their surface density is comparable to the dislocation density of the GaN layer in the order of 10^8–10^9 cm^{-2}. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

Quantum wells (QWs) of $In_xGa_{1-x}N$, embedded into GaN, form the active region in recently developed short-wavelength light emitting and laser diodes [1]. At high x_{In}-values, these quantum wells grow under conditions of high compressive strain. Also, thermodynamic calculations predict the existence of an immiscibility gap in $In_xGa_{1-x}N$ [2]. Both conditions can cause growth mode instabilities. Thus, a three-dimensional (3D) growth resulting in quantum well thickness fluctuations or Stranski–Krastanov islands as well as phase-separation into In-rich and In-poor regions may occur. This behaviour could explain the observation of light emission originating from quantum dot-like localized states within the InGaN quantum wells [3,4].

The growth mode will also be determined by the growth conditions. In general, due to the rather high growth temperatures around 1050°C for GaN and 800°C for InGaN in MOVPE, a step-flow growth mode will be favoured. However, due to the high density of defects, commonly in the order of more than 5×10^8 cm^{-2}, the step-flow mode has to compete with a defect-induced nucleation mechanism. The observed defects in GaN(0 0 0 1) layers on (0 0 0 1) sapphire substrates are mainly

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threading dislocations of either pure edge, mixed, or pure screw character [5]. Threading dislocations having a component of the diplacement vector normal to the crystal face at which they emerge (dislocations of partial or pure screw character) can send out successive turns of steps (in form of spirals) leading to the growth of pyramids [6]. This nucleation mechanism becomes of particular importance in the case of exactly oriented substrate surfaces with low intrinsic step density as well as in the case of "step-blocking" due to the accumulation of impurities at the lateral growth front.

In the following, we will report on the enhanced formation of InGaN spiral islands after deposition of Si onto the GaN surface prior to growth of InGaN. A similar procedure was described to produce self-assembled dots of GaN on a surface of AlGaN [7]. We will show that structures with buried spiral islands exhibit enhanced luminescence efficiency and an increased radiative recombination lifetime in comparison to quantum wells with more flat interfaces.

2. Experimental procedure

The InGaN structures were grown on 1.8–2 μm thick GaN on *c*-plane sapphire films by metal-organic chemical vapor deposition (MOCVD) using the precursors trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia. Disilane was used as n-type dopant. The growth temperature (T_{gr}) of the GaN : Si base layer was 1060°C [8], the InGaN growth temperature 790°C. During InGaN growth, the TMGa flow was 5 μmol/min, the TMIn flow 14 μmol/min, and the ammonia flow 0.35 mol/min, at a total gas flow of 10.3 l/min. The growth time of the InGaN layers corresponded to a nominal quantum well thickness of 3 nm, the nominal In-composition was 22%. Pronounced formation of spiral islands was observed for samples where the GaN layer was exposed to a disilane flow of 2.2 nmol/min for 14 s prior to InGaN deposition (corresponding to a coverage of about 0.01 ML of Si). In order to measure photoluminescence, some of the samples had been capped with 33 nm GaN after island growth.

The surface morphology had been studied with a Digital Instruments Nanoscope III atomic force microscope (AFM) operated in tapping mode. Photoluminescence (PL) measurements were performed at room temperature using the 325 nm line of a He–Cd laser (excitation density 250 mW/cm^2). For the time-resolved PL, a tunable Ti : Sapphire laser operating at a repetition rate of 80 MHz was used.

3. Results and discussion

Fig. 1a shows the typical surface morphology of a (0 0 0 1) GaN surface ($T_{\mathrm{gr}} = 1060$°C). The surface is characterized by almost equally spaced steps of the height of one GaN-monolayer. Some of the steps terminate at threading dislocations with pure or partial screw character [5]. The step length decreases and the surface becomes more rough after deposition of 3 nm InGaN at 790°C (Fig. 1c). This is a natural consequence of the lower diffusivity of adatoms at lower growth temperatures. But already under these conditions, spiral growth around dislocations starts to compete with the step-flow mechanism (Fig. 1c). When the GaN surface steps have been "poisoned" by a preflow of disilane, predominantly spiral growth is observed (Fig. 1b). In this case, the entire surface becomes covered with InGaN islands of a rather uniform size, with a density corresponding to the density of dislocations with pure or partial screw character emerging the surface.

Fig. 2. shows a magnification of a typical spiral island. It is a double-spiral with two parallel turns due to the development of monolayer height terraces. This results from the nature of screw dislocations in a Wurtzite structure having a Burgers vector $b = c$, whereby 1 monolayer $= c/2$. Fig. 2 also shows that these spiral islands are rather flat extended objects, with a base width in the order of 200 nm. The height of this particular island amounts to 1.3 nm (= 5 monolayers). The dark spot in the center of the island is the emerging point of a threading dislocation with screw character. Two further dark pits in the neighborhood, developing no spirals, are most probably emerging points of pure edge dislocations. Besides the growth

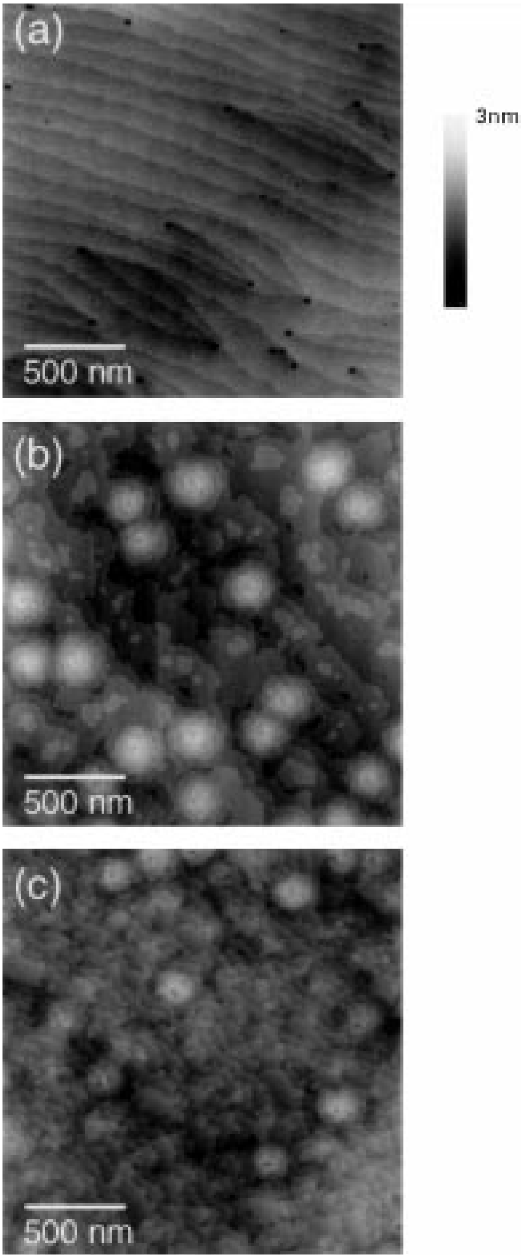

Fig. 1. $2 \times 2\,\mu m^2$ AFM images of (a) GaN ($T_{gr} = 1060°C$), (b) GaN/Si/InGaN ($T_{gr} = 790°C$) and (c) GaN/InGaN ($T_{gr} = 790°C$).

spiral, a few 2D islands exist directly on the terraces, with no links to the next higher monolayer step. This behavior is typical for surface regions where the dislocation density is too low to provide the necessary number of nucleation sites in order to decrease the supersaturation in the vapor.

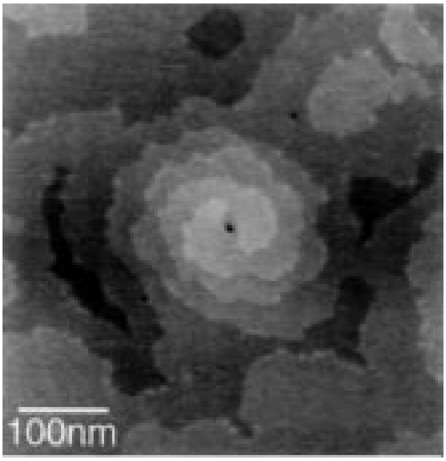

Fig. 2. $500 \times 500\,nm^2$ AFM image of an area with an individual island (note the double spiral), 5 ML in height. The black spot at the top is the emerging point of a screw dislocation. Two other black spots, not surrounded by growth spirals, are most probably due to dislocations of pure edge character.

The dopant Si is known to inhibit step-propagation also on other III–V surfaces. We have shown that with increasing Si-doping the deposition rate in InGaN/GaN quantum well structures decreases [9]. We see also, that in case of the InGaN spiral islands, the amount of deposited InGaN material is much less than predicted by the nominal growth rate (nominal QW thickness: 3 nm, height of the observed islands: < 2 nm). Therefore, we believe that the effect of Si in case of the island growth, discussed in this study, is less related to silicon-induced changes in the surface energies [7] but rather to step blocking due to an accumulation of Si at step and kink surface sites. A very simple atomistic explanation of the growth-inhibiting effect of Si can be given by considering that Si replaces Ga-atoms. A Ga-terminated surface is ready to bind the lone-pair electron of NH_3 at steps in [1 1 2 0] direction. In those cases where Ga-atoms are substituted by Si, there will be an activation barrier to remove the excess electrons (which most probably still bind H-atoms) in order to form Si–N bonds. This activation energy could be expected in the order of 318 kJ/mol (the energy of a Si–H

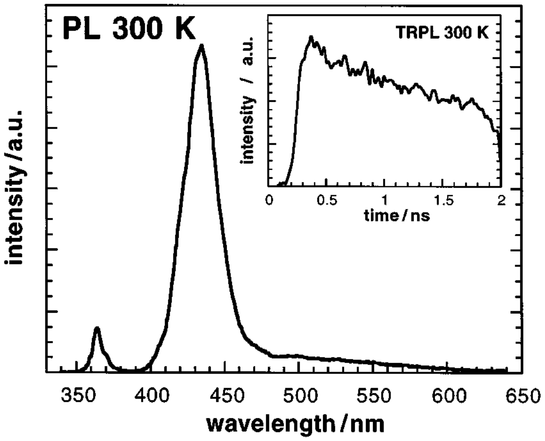

Fig. 3. RT PL spectrum of a structure with buried InGaN spiral islands grown after deposition of about 0.01 ML of Si onto the GaN surface. The inset shows the RT TRPL spectrum.

bond). Assuming the Si sticks at these step positions, it will only inhibit the intrinsic step motion, whereas it will have no effect on the spiral step development around the dislocation center. As a consequence, the InGaN will crystallize around the dislocations under conditions which permit deposition of high quality and In-rich material due to the possibility of lateral strain relaxation, lowering the strain restrictions for an efficient incorporation of In.

The 300 K PL spectra and the results of time-dependent measurements of capped island samples are shown in Fig. 3. The luminescence intensity is significantly higher and the radiative recombination lifetime of $\tau = 1.5$ ns is considerably longer than in the case of conventional single quantum wells ($\tau = 160$–300 ps) [10]. The increase in the PL intensity can be partially explained by the increased electron concentration in the InGaN conduction band due to carrier transfer from the highly doped GaN barrier. However, the drastic increase in the PL lifetime is very similar to observations made in the case of InGaN multi quantum well structures which has been related to dot-like localized states [3,4].

4. Conclusions

An intentionally performed growth mode transition from step-flow into a 3D spiral growth mode by predeposition of Si prior to InGaN growth enhances the PL efficiency and increases the radiative PL recombination lifetimes of InGaN/GaN structures. We attribute this effect mainly to the improved InGaN growth conditions around screw dislocations, where a partial lateral strain relaxation during InGaN deposition becomes possible.

Acknowledgements

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References

[1] S. Nakamura, G. Fasol, The Blue Laser Diode, Springer, Berlin, 1997.

[2] I.-H. Ho, G.B. Stringfellow, Appl. Phys. Lett. 69 (1996) 2701.

[3] S. Chichibu, T. Azuhata, T. Sota, S. Nakamura, Appl. Phys. Lett. 70 (1997) 2822.

[4] Y. Narukawa, Y. Kawakami, M. Funato, S. Fujita, S. Nakamura, Appl. Phys. Lett. 70 (1997) 981.

[5] X.H. Wu, L.M. Brown, D. Kapolnek, S. Keller, B. Keller, S.P. DenBaars, J.S. Speck, J. Appl. Phys. 80 (1996) 3228.

[6] W.K. Burton, N. Cabrera, F.C. Frank, Philosoph. Trans. Roy. Soc. A 243 (1951) 299.

[7] S. Tanaka, S. Iwai, Y. Aoyagi, Appl. Phys. Lett. 69 (1996) 4096.

[8] B.P. Keller, S. Keller, D. Kapolnek, W.-N. Jiang, Y.-F. Wu, H. Masui, X.H. Wu, B. Heying, J.S. Speck, U.K. Mishra, S.P. DenBaars, J. Electron. Mater. 24 (1995) 1707.

[9] S. Keller, A.C. Abare, M.S. Minsky, X.H. Wu, M.P. Mack, J.S. Speck, E. Hu, L.A. Coldren, U.K. Mishra, S.P. DenBaars, Proc. Int. Conf. SiC, III-Nitrides and Related Materials, Stockholm, Sweden, 1997, Mater. Sci. Forum 264–268 (1998) 1157.

[10] C.-K. Sun, S. Keller, S. Wang, M.S. Minsky, S.P. DenBaars, J.E. Bowers, Appl. Phys. Lett. 69 (1996) 1936.