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Growth optimization for thick crack-free GaN layers on sapphire with HVPE

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Conditions for optimized growth of thick GaN layers with crack-free surfaces by HVPE are reported. It was found that a 1:1 mixture of H_2/N_2 as carrier gas leads to the lowest density of cracks in the surface. Crack formation also depends on the properties of the GaN/sapphire templates used. Best results have been obtained for 5 µm thick GaN/sapphire templates grown by MOVPE with medium compressive strain ε_{zz} of about 0.05%. But there is no simple dependence of the crack formation on the strain status of the starting layer indicating that the HVPE growth of GaN can itself introduce strong tensile strain.

1 Introduction

Hydride Vapor Phase Epitaxy (HVPE) has a high potential for the growth of thick GaN layers [1] that can be used as substrates for the subsequent growth of device layer structures using MOVPE or MBE. But also in HVPE, GaN growth has commonly to start on a foreign substrate, e.g. sapphire. Initially, a GaCl-pretreatment or a ZnO layer [1] were used to aid nucleation. Later low-temperature nucleation processes were established for direct growth on sapphire in HVPE [2]. However, lattice mismatch and thermal expansion differences result usually in cracks and wafer bending in thick GaN layers. Most cracks run along the {10-10} planes of GaN forming arrays rotated by 120° to each other. The formation and propagation of cracks and the mechanisms of crack healing in subsequently grown GaN layers have been analyzed in [3]. The nucleation process itself is assumed to be the origin of the strong tensile strain during growth resulting in crack formation. These cracks can be healed out with increasing layer thickness if the surface mobility of the reacting species is sufficient. However, buried microcracks remain in the sapphire and they are held responsible for later wafer breakage. The aim of this study is to examine whether the crack formation can be avoided with lower tensile strain in the starting layer. To this end, optimized growth conditions for crack healing in thick GaN layers were established for the reactor used. Then, starting layers with different strain are used to examine the conclusion drawn from [3] that the strain of the starting layer determines the crack formation.

2 Experimental

The growth experiments have been performed in a horizontal AIX-HVPE reactor with optimized flow geometry described elsewhere [4]. The gas flow rates were adjusted to achieve mixing of NH_3 and GaCl on the substrate without vortex formation. The total flow rate of the $NH_3/N_2/H_2$ mixture passing the NH_3 inlet was fixed to 1.5 l/min. Total flow rates of $GaCl/N_2$ and N_2 via the showerheads were 400 ml/min.

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The main gas flow purging the complete reactor was 9, 7, and 5 l/min for 800, 500, and 200 hPa to ensure similar flow patterns above the substrate. Growth was performed at about 1010 °C with variation of the hydrogen fraction, the HCl flow with constant V/III ratio, and the V/III ratio with constant HCl flow on one type of MOVPE-grown templates. Selected growth conditions have been applied to different GaN/sapphire templates grown by MOVPE or by HVPE [4] for the comparison of crack formation. The 2 inch templates were cut into quarters and were overgrown in parallel to ensure identical growth conditions on all templates.

3 Results and discussion

In Fig. 1 the dependence of the growth rate on hydrogen content in the carrier gas for different total pressures is shown. The growth rate decreases with increasing H_2/N_2 fraction almost linearly, it also decreases with decreasing total pressure. Figure 2 shows the dependence of the growth rate on V/III ratio. Above a critical value of around 10 the rate linearly increases with V/III ratio while below this value it rapidly decreases and even etching of the template occurs at very low V/III ratio. All GaN layers from Fig. 1 were found transparent and the surfaces appeared mirror-like with exception of a dull one grown at 800 hPa with pure nitrogen as carrier gas. Most wafers with these thick GaN layers broke due to strong convex bending during or after the cool-down process. One of two 2 inch wafers with a GaN layer of 84 µm thickness grown at 800 hPa with 55% H_2 was found long-term stable despite a curvature radius of only 0.5 m determined by stylus profiler Tencor P-10. This indicates that microcracks which where found by cross-sectional SEM inspection quite similar to those described in [3] are not the only reason for wafer breakage but subsurface damage or scratches in the sapphire may play a role.

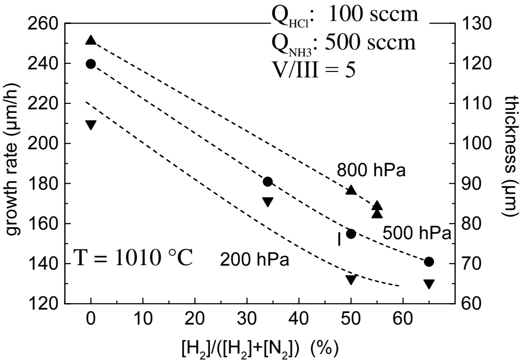

Fig. 1 Growth rate in dependence on hydrogen content in carrier gas for different total pressures. Dashed lines are guides to the eye.

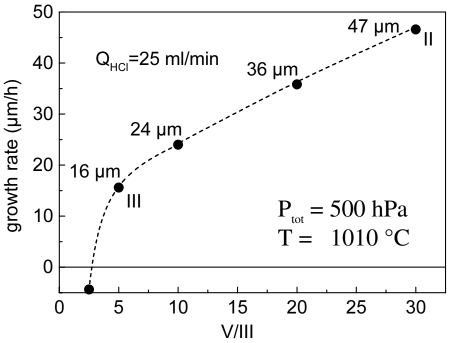

Fig. 2 Growth rate vs. V/III ratio. The negative rate at very low V/III ratio indicate etching of template. Dashed lines are guides to the eye.

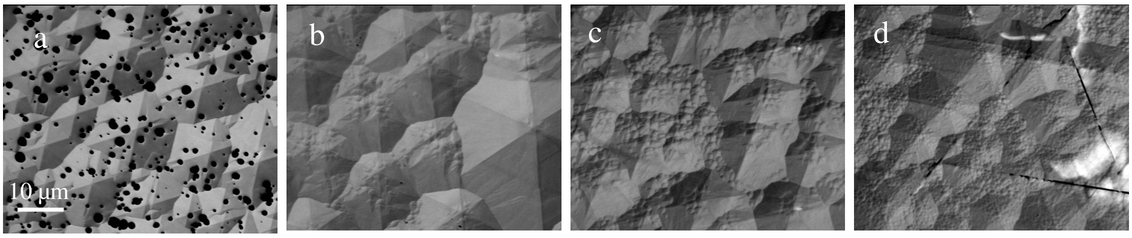

Fig. 3 Optical Normarski micrographs of surface morphology for samples from Fig. 1 grown at 500 hPa and different H_2 fractions in the nitrogen carrier gases, i.e. 0% (a), 35% (b), 50% (c), and 65% (d).

In Fig. 3 the morphology of the GaN layers grown at 500 hPa is shown for different hydrogen fractions in the carrier gas. When the focus of the microscope is moved below the surface the layers show networks of crack arrays twisted by 120°. In addition, layers grown with pure nitrogen in the carrier gas show a considerable mud cracking at the surface which is probably formed during the cool-down process. The etch pits observed at the surface of Fig. 3a grown with pure nitrogen carrier gas are due to insufficient surface stabilization. The etch pits vanish for lower total pressures. At 800 hPa a high pit density results in a dull surface.. The pits also vanish when hydrogen is mixed to the carrier gas. Adding hydrogen smoothes the surface morphology by reducing the large crystal facets observed in Fig. 3a due to surface kinetics [5]. The surface of the layer grown with 50% hydrogen is completely crack-free (Fig. 3c), whereas numerous open cracks are observed for 65% hydrogen (Fig. 3d). The surfaces of the layers grown at 800 hPa and 200 hPa show similar features. PL investigations of the surface strain ε_{zz} revealed a minimum of strain for growth with 50% hydrogen. All layers show dislocation densities of about (2±1) x 10^8 cm^{-2}. From these findings we deduced that 50% H_2 in the carrier gas represents an optimum for growth.This also holds for the background doping. Further growth investigations were thus restricted to 500 hPa total pressure and 50% hydrogen. The flow rate of HCl was varied from 100 ml/min to 25 ml/min using a constant V/III ratio of 5 and the V/III ratio was varied for constant HCl flow (Fig. 2). All

Table 1 Overview on the different templates characterized by XRD, PL, and surface profiling.

Template	Ω 00.2 (arcsec)	Ω 01.5 (arcsec)	ε_{zz} (HRXRD) (%)	ε_{zz} (PL) (%)	R (P-10) (m)
A (1.5 µm)	94	131	0.12	0.125	–5
B (4.9 µm)	266	153	0.045	0.050	–6
C (2.9 µm)	320	173	0.041	0.054	–9
D (3.6 µm)	626	308	–0.008	–0.009	–40

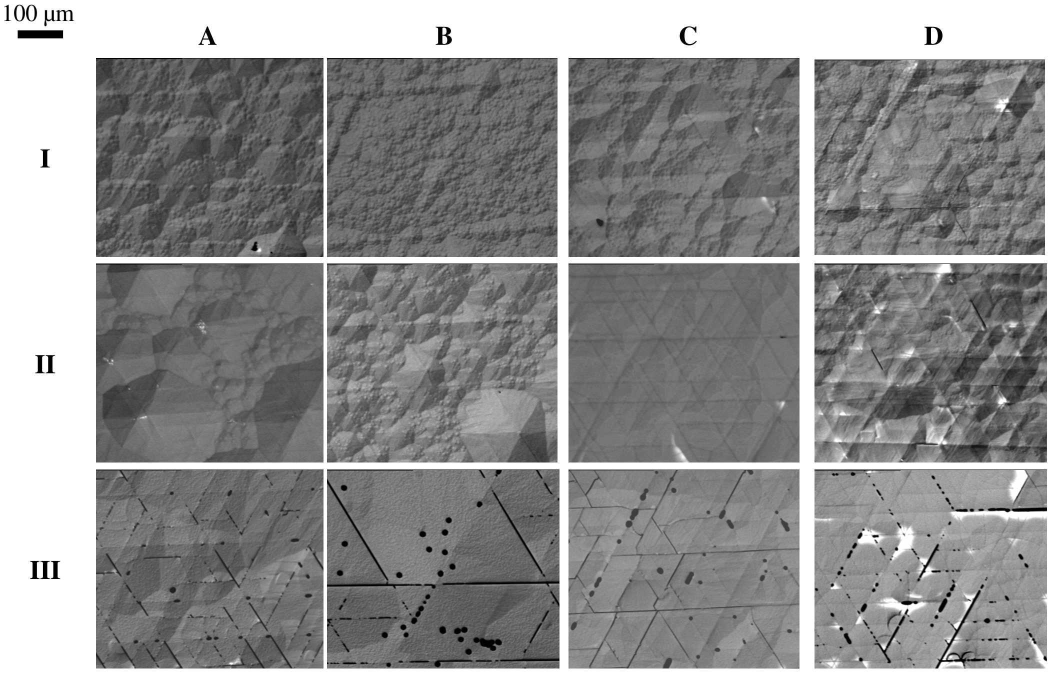

Fig. 4 Conditions **I**: V/III=5, Q_{HCl}=100 ml/min, d=35 µm; **II**: V/III=30, Q_{HCl}=25 ml/min, d=35 µm; **III** : V/III=5, Q_{HCl}=25 ml/min, d=15 µm.

layers above the lowest V/III ratio of 2.5 are mirror-like and transparent. The surface morphology observed for the GaN layers grown with these different conditions are similar to the one shown in Fig 3c. Wafers with GaN layers of thicknesses below 35 µm did not break but open cracks are still observed on their surfaces.

Strain built in during the nucleation process on sapphire was discussed as reason for the crack formation during subsequent HVPE growth in the GaN layer and the sapphire substrate [3]. To check this explanation three different growth conditions (see Fig. 4 details in figure caption) have been applied to different GaN/sapphire templates A, B, C from MOVPE growth, and D from HVPE growth (Tab. 1). These templates cover a wide range of strain at the GaN surface from heavily compressive (A) over medium compressive (B,C) typical for many MOVPE-grown layers to slightly tensile strain (D) for the HVPE-grown template similar to [6]. The thickness of the layers was about 35 µm (reduced to 15 µm thickness for condition III).

Comparing the samples B and C which are similar in strain and bending values reveals remarkable differences. For condition III the spacings between the cracks of each array are larger for sample B than for sample C. The spacing value can be used to estimate the tensile film stress [3] which should therefore be larger in sample C than in sample B. For condition I and II the surface of sample B is already closed, i.e. the cracks are healed out whereas the surface of sample C shows still a few open cracks. In addition, in sample C bright spots due to spontaneous delamination of the layer are observed. Indeed, strain determination of the surface by PL measurements according to [7] revealed compressive strain ε_{zz} of 0.036% and 0.022% for sample B but tensile strain ε_{zz} for sample C of -0.002% and -0.003% for the conditions I and II. It is concluded that different templates which may be comparable in their surface strain after MOVPE growth can induce different crack patterns and different strain into HVPE-grown GaN layers. The surface morphology of sample A appears much better than the one of sample D for conditions I and II. But there are still small bright spots indicating a tendency for local layer delamination. For condition III rather similar crack distances arefound for samples A, C, and D. Partial layer delamination and open cracks are found for sample D under all conditions. These results indicate that there is no or at least no simple dependence of the crack formation on the strain of starting templates.

4 Conclusions

A mixture of H_2/N_2 in the carrier gas of about 1:1 was found to be optimum for the growth of thick GaN layers with crack free surfaces grown by HVPE at different total pressures. Using appropriate GaN/sapphire starting layers the healing of cracks which occurs in an initial stage of growth due to tensile strain was found to be completed after more than 35 µm with strain-free surfaces. GaN/sapphire templates within a strain range from heavily compressive to slightly tensile were evaluated with respect to their influence on crack formation. Tensile-strained starting layers were found to yield the worst results. Best results have been achieved with thick GaN templates grown by MOVPE. But starting layers with similar strain status can lead to different crack formation. In summary, there is no straightforward dependence of crack formation on the strain of a template as might be deduced from earlier analysis [3]. This indicates that the used HVPE growth process introduced tensile strain itself. Hence, growth conditions for less tensile strain have to be found to avoid crack formation.

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