Vertical-HVPE as a Production Method for Free-Standing GaN-Substrates

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Abstract

The cost effective growth of free standing GaN substrates requires the boule growth approach. We developed a Vertical Hydride Vapor Phase Epitaxy (VHVPE) tool for the growth of up to 7 cm long GaN boules of 2 inch size. Prototypes were built and optimized with respect to total growth rate, parasitic ammonium chloride deposition and growth uniformity of the boule.

INTRODUCTION

GaN and its related alloys have opened up the road to efficient solid state lighting by white light emitting diodes (LED) and are beginning to revolutionize the field of high capacity data storage by the blue laser diode (LD). However, the growth on non-native substrates such as sapphire or SiC requires elaborate schemes of defect reduction to increase the luminous efficacy of the devices by preventing non-radiative recombination paths along dislocations. The native GaN substrate, however, remains elusive: The high pressure solution method [1], ammonothermal growth [2], physical vapor transport (PVT) [3] or sublimation growth [4] still face the challenges of irregular growth of boules or scalability of size. Hydride-Vapor-Phase Epitaxy (HVPE) on the other hand offers a reasonably high growth rate and a controlled geometrical size, yet as an epitaxial method it is usually limited to single wafer production. The mass production of free-standing GaN substrates can be facilitated by the expansion of the well known horizontal HVPE system to a multi-wafer HVPE approach or the growth of GaN boules in a vertical system.

DESCRIPTION OF THE GROWTH SYSTEM

A sketch of the reactor layout is shown in Fig. 1. The growing boule is placed upside-down above the gas injector head. A retraction unit allows an in-situ adjustment of the optimal distance between growth surface and gas injector and facilitates to maintain this optimal distance constant as the boule grows longer by pulling it up with the speed of growth.

Two independently controllable radio-frequency (RF) heating zones provide accurate temperature control along the entire length of the reaction cell. The lower heating zone being capable to control the temperature between room temperature (RT) and 900°C serves as the heat source for the liquid Ga source. The upper RF heater provides growth temperatures between RT and 1200°C at the reaction zone. This two RF heater design with reduced thermal masses in the graphite ring susceptors allows to independently cool down the upper zone for boule harvesting while maintaining the lower zone and the Ga source near operating temperature.

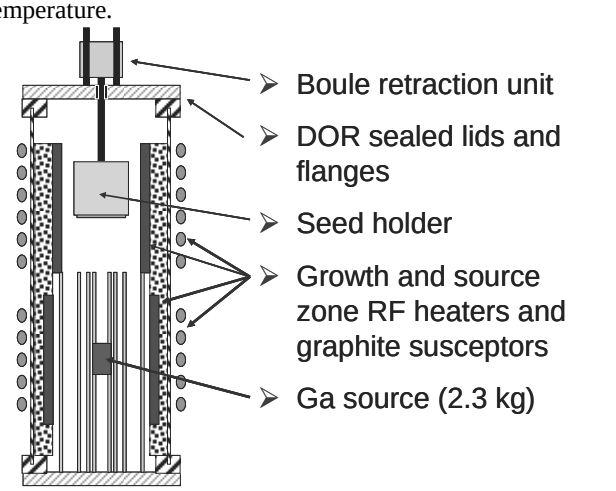

Fig. 1: Schematic sketch of the VHVPE growth reactor detailing its components (not drawn to scale).

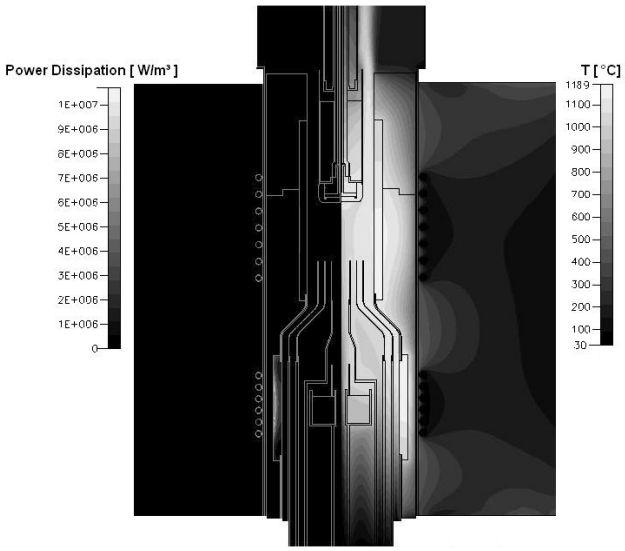

Fig. 2: Numerical simulation of the temperature distribution in the reactor. Power dissipation (left) and temperature distribution (right) are shown (not drawn to scale).

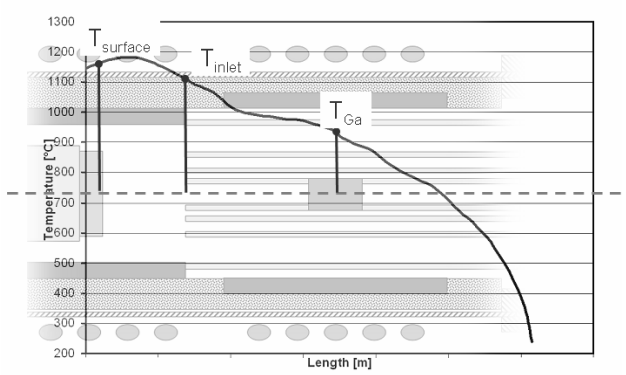

Fig. 3. Temperature scan along the reactor axis (from numerical simulation).

Fig. 2 shows numerical simulations of the temperature distribution under normal operating conditions, i.e. source and growth temperatures at T_{Ga} = 850°C and $T_{surface}$ = 1100°C, respectively, and gas flow included. As can be seen the RF power is completely dissipated in the graphite heater elements. Thus, no RF power is induced into the reactor interior rendering the setup a true hot wall reactor. Consequentially, the entire volume of the growth zone is kept at the same temperature and heat transport from the seed holder through the thick boule to the growth surface is not necessary to maintain the growth temperature at the surface.

Fig. 3 shows a temperature scan along the reactor center axis. As can be seen all temperatures downstream of the Ga source up to the growth surface are higher than T_{Ga} avoiding condensation of GaCl at the reactor components.

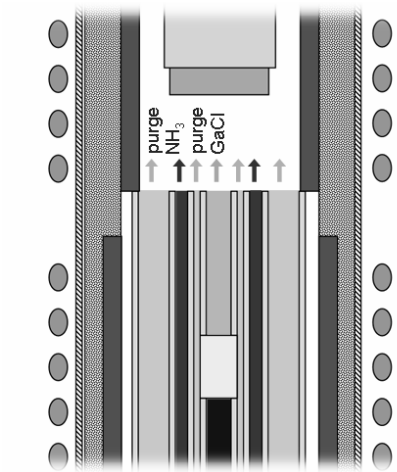

Fig. 4: Concentric reactor inlet geometry.

Fig. 4 shows a cut through the inlet design. All gas lines of the injector head are concentric. A sheath flow between the reactive species GaCl and NH_3 provides separation under laminar flow conditions and, thus, avoids pre-reactions. Complete mixing of the species is facilitated in the stagnation zone right before the growth surface, effectively mixing the species and providing a spatially uniform V/III ratio.

HVPE of GaN is challenging to the design of the growth reactor since at temperatures below 350°C HCl and NH_3 form ammonium-chloride (NH_4Cl), a white powder that can create growth defects and could lead to blockage of the exhaust lines. Since both HCl and NH_3 are present in the exhaust stream, the formation of ammonium-chloride somewhere in the exhaust area is unavoidable. Therefore, advanced designs are necessary to effectively lead the hot gases out of the reactor cell and into the filter trap, avoiding cold spots along the way.

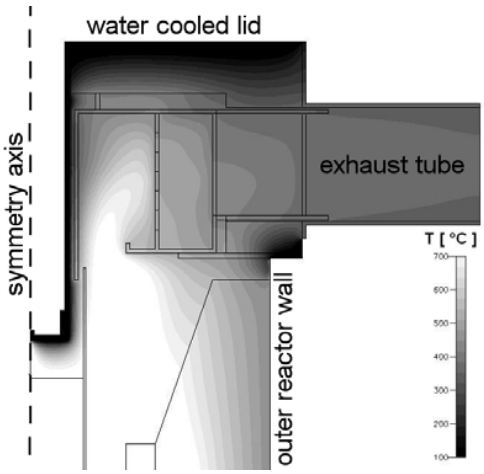

Fig. 5: Numerical simulation of the exhaust collector temperature distribution under process conditions (cross section not drawn to scale).

Fig. 5 shows a simplified result of a numerical simulation of the reactor exhaust area. As can be seen the entire exhaust area with its pressure manifold is comfortably above 350°C. Hence, the formation of ammonium-chloride inside the reactor cell under normal operating conditions is avoided.

In-situ metrology has become a powerful tool for the optimization and monitoring of growth processes. Therefore, an optical access to the substrate was integrated along the reactor main axis. This allows the monitoring of Fabry-Perot interferences, reflectance and the pyrometric measurement of the surface temperature at the center of the growing boule.

Experimental Results

With these design guidelines prototype systems were built and evaluated with respect to their performance for the growth of GaN boules.

Fig. 6 shows a white light interference (WLI) thickness map of a layer grown on a sapphire/GaN/SiN_x-hole ELOG mask template. The measured thickness corresponds to a growth rate of ~400 µm/h with a standard deviation of 10.2% for 2 inch diameter. This result was achieved at an H_2/N_2-carrier gas ratio of unity at the surface, since

hydrogen is commonly used as a surfactant to improve morphology at the price of a reduction in growth rate.

With these process conditions 1.4 mm thick layers could already be grown. However, crack formation due to the build-up of strain for layers grown on seeds with non-native carrier substrates remains a challenging issue which is still subject to investigation. For the time being, therefore, lower growth rates have proven to yield better morphology. Fig. 7 shows a 130 µm thick layer grown at a moderate growth rate of 250 µm/h. As can be seen the surface morphology is specular and no crack formation is observed.

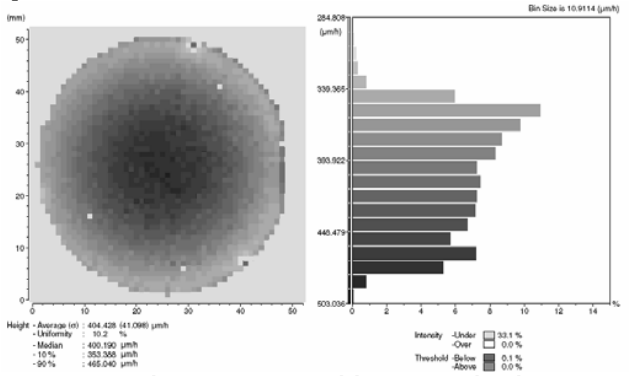

Fig. 6: WLI growth rate map over a 2 inch layer. 400 µm/h with a uniformity of 10.2% were achieved.

However, transferring these successes to thicker layers remains challenging since changes in the morphology of the surface are observed. Fig. 8 shows AFM scans of two layers grown under identical growth conditions. As can be seen the thin layer features neat parallel steps of 10-15 µm height spaced at 5 nm.

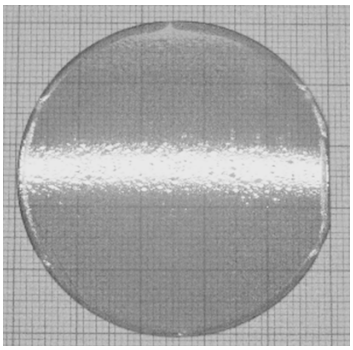

Fig. 7: Photograph of a 130 µm thick layer grown at 250 µm/h.

As the layers become thicker a transition in the surface morphology is observed. For a 2 mm thick layer the step-flow growth disappears and the surface becomes irregular with pits of about the same depth (10-15 nm). Fortunately, the overall RMS roughness does not increase. It was measured to be 3 nm for a 10x10 µm^2 scan field [5,6].

The etch pit density (EPD) of the layers decreased from 2x10^6 cm^{-2} for the 250 µm thin layer to 5x10^5 cm^{-2} for the 2 mm thick layer [5,6]. This indicates that the dislocation associated with the etch pits also decrease in density for thicker layers.

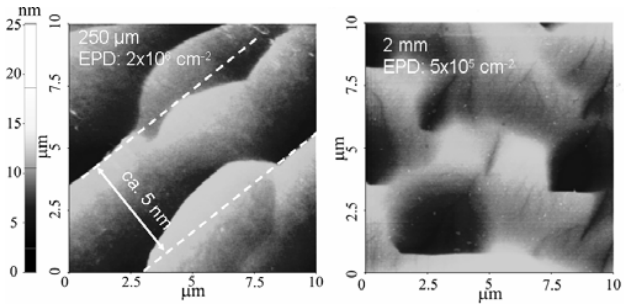

Fig. 8: AFM scans of 10x10 µm areas of a 250 µm (left) and a 2 mm (right) thick layer. Height scale identical in both pictures.

The overall crystalline quality is also reflected in low temperature (LT) photoluminescence (PL) measurements recorded at 2 K [5,6]. Fig. 9 shows that excitons up to the free-B (FEB) exciton can be observed. The spectrum is dominated by the donor-bound exciton (DBE). This, in combination with the also visible acceptor-bound exciton (ABE) and the well resolved donor-acceptor pair (DAP) transitions and their LO-phonon replica indicates the presence of small amounts of Si and O in the layers. The acceptor is believed to be Mg proliferated from the upper layer of an LED structure which was used as the seed template for this layer.

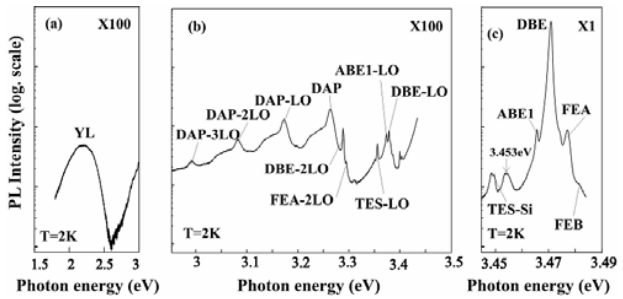

Fig. 9: LT-PL measurements of the yellow (left), DAP (center) and exciton (right) spectral ranges. Attention: different scales!

CONCLUSIONS

These results show that the growth of thick GaN layers in a VHVPE system is feasible. Growth rates as high as 400 µm/h were demonstrated under generic process conditions with hydrogen present at the growth surface. Up to 2 mm thick layers were grown and characterized. The observed change in the growth morphology from step-flow to valley-like did not impair the overall crystalline quality as the observed EPD was reduced indicating a reduction in dislocation density as the layer grows thicker and the RMS roughness for a 10x10 µm^2 area scan field remained constant at 10-15 nm. Additionally, such layers showed LT-PL spectra with resolved B-excitons. However, additional effort in the development of the growth processes is necessary, before boules with up to 7 cm length can be grown.

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ACRONYMS

ABE: Acceptor Bound Exciton
AFM: Atomic Force Microscopy
DAP: Donor-Acceptor Pair
DBE: Donor Bound Exciton
ELOG: Epitaxial Lateral Overgrowth
EPD: Etch Pit Density
FEA: Free A-Exciton
FEB: Free B-Exciton
HVPE: Hydride Vapor Phase Epitaxy
LD: Laser Diode
LED: Light Emitting Diode
LO: Longitudinal Optical
LT: Low Temperature
PL: Photoluminescence
PVT: Physical Vapor Transport
RF: Radio Frequency
RT: Room Temperature
VHVPE: Vertical HVPE
WLI: White Light Interference
YL: Yellow Luminescence