HVPE-grown free-standing GaN of high structural and optical quality

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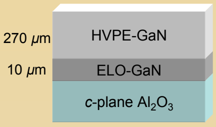
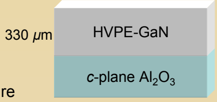
→ Outline

- We demonstrate the growth of high-quality and stress-free **bulk-like GaN** by **hydride vapor phase epitaxy** (HVPE) in a vertical atmospheric-pressure reactor.
- The GaN layers with thicknesses up to 330 μm were either grown directly on sapphire substrate or using a two-step epitaxial lateral overgrown GaN template on sapphire.
- XRD and PL data prove the **high crystalline quality** of the free-standing material. The dislocation density as inferred from plan-view TEM images is 1–3×10^{7} cm^{-2}.
- The main near-band gap PL emission lines and the phonon spectra obtained from IR ellipsometry and Raman measurements show that the material is **virtually stress-free** close to the Ga-face.
- The GaN material presented here is well suited to **serve as a lattice- and thermal-expansion-coefficient matched substrate** for further homoepitaxial growth, as needed for high-quality III-nitride device applications.

→ Sample preparation

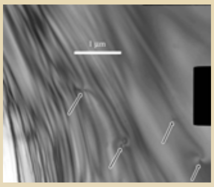
- ► Hydride vapor phase epitaxy growth at Linköping University
- ► New type of vertical HVPE growth reactor with a bottom-fed design
- ► Growth either directly on *c*-plane sapphire substrate or using a ~10 μm-thick two-step ELO GaN template on sapphire substrate (LUMILOG, France)
- ► Growth temperature = 1090 °C;
- ► V:III ratio = 12 – 24
- ► Average growth rate = 70 – 110 μm/h
- ► Delamination from the substrate by a laser-induced lift-off process or self-separation from the substrate upon cooling down to room temperature





→ Structural characteristics

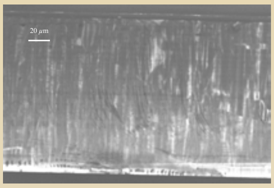
TEM



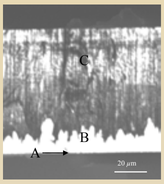
Plan-view TEM image of the as-grown Ga-face (sample on ELO template). The dislocations intersecting the surface are marked by arrows.

Dislocation density (on the Ga-face): 1–3×10^{7} cm^{-2}

CL

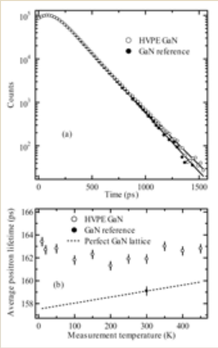


Panchromatic CL image of the HVPE-GaN layer grown on ELO template, revealing homogeneous high-quality material.



Typical panchromatic CL image of a HVPE-GaN layer grown directly on sapphire, revealing three different structural areas.

e^{+} annihilation

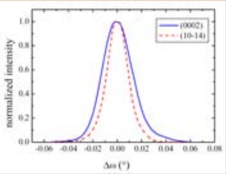


Ga vacancy related defect concentration: 5–8×10^{15} cm^{-3}

(a) Positron lifetime spectra at 300 K measured in the HVPE-GaN sample on ELO template and a GaN reference sample, where no positron trapping at vacancies is observed. The solid lines are fits of exponential decay components to the data.

(b) The positron lifetime measured in the HVPE-GaN sample on ELO template as a function of measurement temperature. The dashed line describes the effect of the thermal expansion on the positron lifetime in the GaN lattice observed in bulk GaN:Mg.

XRD



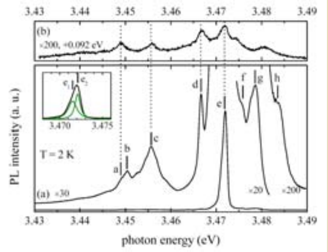
X-ray rocking curves of different reflections, measured on the Ga-face of the HVPE-GaN layer grown directly on sapphire:

FWHM values (slit width 1 mm):

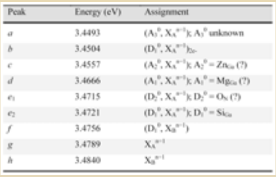
reflection	ω-scans	2θ-ω scans
(1 0 –1 4)	96 arcsec	143 arcsec
(0 0 0 2)	129 arcsec	58 arcsec

→ Optical characteristics

PL



(a) Near-band gap PL spectrum taken on the Ga-face of the GaN layer on ELO template. The inset gives a detailed view on peak *e*, which is actually composed of a doublet structure. (b) PL spectrum recorded in the range of 1-LO phonon replicas of the acceptor bound excitons, shifted by +92 meV for direct comparison to the spectrum in (a).



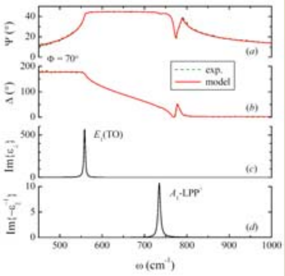
Peak	Energy (eV)	Assignment
a	3.4493	$(A_3^0, X_A^{n=1})$: A_3^0 unknown
b	3.4504	$(D_1^0, X_A^{n=1})_{2e}$
c	3.4557	$(A_2^0, X_A^{n=1})$: $A_2^0 = Zn_{Ga}$ (?)
d	3.4666	$(A_1^0, X_A^{n=1})$: $A_1^0 = Mg_{Ga}$ (?)
e$_1$	3.4715	$(D_2^0, X_A^{n=1})$: $D_2^0 = O_N$ (?)
e$_2$	3.4721	$(D_1^0, X_A^{n=1})$: $D_1^0 = Si_{Ga}$
f	3.4756	$(D_1^0, X_B^{n=1})$
g	3.4789	$X_A^{n=1}$
h	3.4840	$X_B^{n=1}$

Spectral features detected in the near-band gap PL spectrum and their most plausible assignments.
For the N-face, the *e*$_1$ and *e*$_2$ emission lines are 0.3 meV below those of the Ga-face.

The line positions of the main donor bound exciton emissions (*e*$_1$ and *e*$_2$) suggest *virtually strain-free* material on both surfaces.

The small full width at half maximum values of the donor bound exciton lines (sample on ELO template: *e*$_1$=1.19 meV and *e*$_2$=0.71 meV for the Ga-face; *e*$_1$=1.53 meV and *e*$_2$=1.21 meV for the N-face) indicate *high crystalline quality of both surfaces*.

IR ellipsometry



The *E*$_1$(TO) phonon position measured at 558.52 cm^{-1} (Ga-face) confirms the small residual stress in the material (σ = 0.12 GPa).

The free-electron concentration is estimated to be less than 2×10^{17} cm^{-3}.

Mid-IR ellipsometric Ψ (a) and Δ (b) spectra taken on the Ga-face of the sample on ELO template.

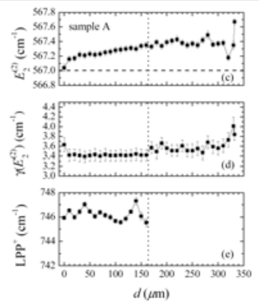
(c) Imaginary part of ε$_\perp$ revealing the *E*$_1$(TO) phonon and (d) imaginary part of the dielectric loss function parallel to the *c*-axis, where the *A*$_1$-LPP$^+$ mode is peaking.

μ-Raman scattering

In the upper ~160 μm of the film directly grown on sapphire, the (compressive) stress does not exceed 0.10 GPa.

At the same time, the crystalline quality reflected by the phonon linewidth remains highly constant.

A widely homogeneous free-electron distribution in this zone is inferred from the vertical LPP$^+$ mode profile.



Frequency and linewidth of the *E*$_2^{(2)}$ phonon mode, and the LPP$^+$ mode frequency vs. distance *d* from the Ga-face (sample grown directly on sapphire).

Related publications:

- A. Kasic *et al.*, "*Micro-Raman scattering profiling studies on HVPE-grown free-standing GaN*", phys. status solidi, ISBLLED-2004 proceedings
- D. Gogova *et al.*, "*Structural and optical properties of bulk-like GaN grown by hydride-vapor phase-epitaxy on two-step epitaxial lateral overgrown GaN*", J. Appl. Phys., submitted
- D. Gogova *et al.*, "*Optical and structural characteristics of free-standing bulk-like GaN*", Jap. J. Appl. Phys., to be published
- D. Gogova *et al.*, "*Characterization of high-quality free-standing GaN grown by HVPE*", Physica scripta, to be published
- H. Larsson *et al.*, "*Free-standing HVPE-GaN layers*", phys. status solidi (c) **0**, 1985 (2003)
- D. Gogova *et al.*, "*Fast growth of high quality GaN*", phys. status solidi (a) **200**, 13 (2003)

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