Mg-doping and free-hole properties of hot-wall MOCVD GaN

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The hot-wall metal-organic chemical vapor deposition (MOCVD), previously shown to enable superior III-nitride material quality and high performance devices, has been explored for Mg doping of GaN. We have investigated the Mg incorporation in a wide doping range (2.45 × 10^{18} \text{ cm}^{-3} \text{ up to } 1.10 \times 10^{20} \text{ cm}^{-3}) and demonstrate GaN:Mg with low background impurity concentrations under optimized growth conditions. Dopant and impurity levels are discussed in view of Ga supersaturation which provides a unified concept to explain the complexity of growth conditions impact on Mg acceptor incorporation and compensation. The results are analysed in relation to the extended defects, revealed by scanning transmission electron microscopy (STEM), X-ray diffraction (XRD), and surface morphology, and in correlation with the electrical properties obtained by Hall effect and capacitance-voltage (C-V) measurements. This allows to establish a comprehensive picture of GaN:Mg growth by hot-wall MOCVD providing guidance for growth parameters optimization depending on the targeted application. We show that substantially lower H concentration as compared to Mg acceptors can be achieved in GaN:Mg without any in-situ or post-growth annealing resulting in p-type conductivity in as-grown material. State-of-the-art p-GaN layers with a low-resistivity and a high free-hole density (0.77 Ω.cm and 8×10^{17} \text{ cm}^{-3}) are obtained after post-growth annealing demonstrating the viability of hot-wall MOCVD for growth of power electronic device structures.

I. INTRODUCTION

Mg doping of GaN is essential for obtaining epitaxial layers with p-type conductivity for GaN-based devices such as light-emitting diodes (LEDs), normally-off high electron mobility transistors (HEMTs), and p-n diodes. Metal-organic chemical vapor deposition (MOCVD) is the technique of choice to grow large-scale GaN-based device structures and p-type doping in MOCVD GaN:Mg with free hole concentrations in the range of 10^{16} - 10^{18} \text{ cm}^{-3} has successfully been demonstrated. [1, 2, 3, 4] Originating from the pyrolysis of hydrogen carrier gas and from the ammonia precursor cracking, atomic H is abundant in the MOCVD processes. The incorporated hydrogen must dissociate from the Mg-H complexes and out-diffuse to render GaN p-type, which requires post-growth treatment of the as-grown material. [5, 6] The activation is typically achieved via ex-situ rapid or regular thermal annealing under N_{2} atmosphere. Due to the large Mg acceptor ionization energy, the free hole concentration at room temperature is much lower than the Mg doping concentration. For this reason, high Mg concentration should be incorporated in GaN to reduce the ionization energy. [1, 7] However, above a certain level, typically in the low 10^{19} \text{ cm}^{-3} range, further increase of Mg concentration leads to a reduction in free hole concentrations. [1, 3, 8] The latter is often associated with the formation of pyramidal inversion domains (PIDs). [2, 8] It has been shown that Mg atoms segregate at the PID (0001) boundaries rendering Mg atoms electrically inactive. [3] Other works have suggested that the nitrogen vacancy, V_{N} and its complexes (e. g. Mg-V_{N}) play a role in the compensation of Mg acceptors at high dopant concentrations. [10, 11] In addition, any donors, e. g. unintentional impurities such as Si and O or native defects such as Ga interstitials, will interfere negatively with the desired p-type conductivity. Notably, C is a major impurity in MOCVD due to the metal-organic precursor molecules cracking. The roles of C as a trap and
compensation defect in GaN:Mg have been extensively discussed. Density functional theory calculations showed that C occupying Ga site, is a shallow donor and it has the lowest formation energy for Fermi level positions close to the valence band maximum. Experimentally, it was demonstrated that the C incorporation leads to an increased donor concentration and a reduction in hole mobility, which was attributed to a donor-like +1/0 state of Cs. Variation of growth conditions, such as V/III ratio, precursor and gas flows, is expected to affect impurity incorporation and defect formation energies, and hence their densities, might influence the degree of passivation/compensation of Mg acceptors. Therefore, optimization and tuning of the growth process is explicitly necessary for acquiring low impurity and native defect levels in GaN:Mg.

Continuous efforts in further improving p-type conductivity and free hole properties, including both growth and post-growth acceptor activation, are undertaken to further the progress in GaN-based device performance. Despite the numerous investigations, no study exists on the capability of hot-wall MOCVD to deliver high-quality GaN:Mg layers with p-type conductivity. Because of its concept and the range of high deposition temperatures (up to 1400 - 1600 °C) achievable, the hot-wall MOCVD has successfully complied to conditions required for deposition of epitaxial layers of Al(Ga)N semiconductor quality and reflected in demonstrating excitation luminescence in photoluminescence measurements and Mg-doped Al$_{0.85}$Ga$_{0.15}$N layers with low resistivity at room temperature. Recently, a room-temperature mobility above 2200 cm$^2$/V.s of two-dimensional electron gas (2DEG) AlGaN/GaN HEMT heterostructures, a GaN–SiC hybrid material for high-frequency and power electronics, and state-of-the-art N-polar AlN epitaxial layers using hot-wall MOCVD have been demonstrated. The hot-wall MOCVD concept enables reduced temperature gradients in both vertical and horizontal directions. It further allows for independent control of the gas phase chemistry over the substrate and the growth species surface diffusion, which may be exploited to control defect formation and impurity incorporation in a wider growth window.

In this work, we report a systematic study of Mg-doped GaN epitaxial layers grown by hot-wall MOCVD. A detailed investigation of the effects of growth and doping conditions on the incorporation of Mg, H and C impurities is presented and discussed in terms of Ga supersaturation. The results are evaluated in relation to extended defects, revealed by transmission electron microscopy as well as X-ray diffraction and surface morphology, and correlated with the electrical and free-hole properties of the GaN:Mg layers obtained by Hall-effect and capacitance-voltage measurements. As a result, a comprehensive picture of hot-wall MOCVD of GaN:Mg is established, demonstrating the capabilities of this technique to deliver state-of-the-art p-type GaN.

II. EXPERIMENTAL DETAILS

A hot-wall MOCVD reactor in horizontal configuration was utilized for the epitaxial growth of all layers. Chemical-mechanical polished 4H-SiC substrates with on-axis (0001) orientation were used after wet chemical cleaning treatment. Before growth, the cleaned substrates were annealed and etched with hydrogen at 1340 °C in the MOCVD reactor. AlN nucleation layers (NL) with thickness of ~50 nm were grown at 1250 °C with a V/III ratio of 1258, followed by growth of 500 nm-thick Mg-doped GaN layers. The growth process was performed under a constant ammonia (NH$_3$) flow (2 l/min), a mixture of N$_2$ and H$_2$ as carrier gases and a constant pressure of 100 mbar. NH$_3$, Trimethylaluminum (TMAI), Trimethylgallium (TMGa) and bis(cyclopentadienyl)magnesium (Cp$_2$Mg) were used as the N, Al, Ga and Mg precursors respectively. The growth conditions of the GaN:Mg layers are summarized in Table I. The unintentionally doped (UID) GaN reference sample and the Mg-doped GaN samples with Mg doping concentration ranging from 2.45×10$^{18}$ cm$^{-3}$ up to 1.10×10$^{20}$ cm$^{-3}$, are denoted as M$_0$ and M$_1$-M$_5$, respectively. Several sets of Mg-doped GaN layers were grown by systematically varying one of the following growth parameters: (i) Cp$_2$Mg/TMGa ratio - samples M$_1$ - M$_5$ grown under optimized conditions with low carrier gas flows, where the Cp$_2$Mg/TMGa is varied between 0.033% and 0.5% and samples - M$_7$ and M$_8$ grown with high carrier gas flows with Cp$_2$Mg/TMGa of 0.335% and 0.5%, respectively, (ii) V/III ratio - samples M$_4$, M$_6$ and M$_7$ grown with Cp$_2$Mg/TMGa of 0.335% and V/III ratios of 906, 1811 and 453, respectively, and (iii) growth temperature (T$_g$) - samples M$_8$ and M$_9$ with T$_g$ = 1120 °C and T$_g$ = 1040 °C, respectively. In addition, a sample, M$_{opt}$ was grown on 1 μm-thick undoped GaN using the conditions of M$_3$. The Mg acceptors were activated using optimized ex-situ annealing process at 900 °C in N$_2$ atmosphere in a rapid thermal processing equipment.

The Mg and the background impurity (H, C, Si, O) depth profiles in as-grown and annealed samples were measured by secondary ion mass spectroscopy (SIMS). The detection limit was 1×10$^{16}$ cm$^{-3}$ for Mg, 1×10$^{17}$ cm$^{-3}$ for H, (3-5)×10$^{15}$ cm$^{-3}$ for Si and O, and 1×10$^{15}$ cm$^{-3}$ for C. The average concentrations for all the elements were estimated using the SIMSView program from EAG Labs. The surface morphology of the layers was studied by atomic force microscopy (AFM) using a Veeco Dimension 3100 scanning probe microscope in tapping mode. Both 5 × 5 μm$^2$ and 80 × 80 μm$^2$ images were acquired. Spectroscopic ellipsometry measurements were performed on a J. A. Woollam RC2-XI ellipsometer in the ultraviolet-visible spectral range (0.7 – 5.9 eV) for the determination of the layer thicknesses. The crystalline quality of the layers was evaluated by high-resolution X-ray diffraction (HRXRD) using a PANalytical Empyrean diffractometer. A hybrid monochromator consisting of a parabolic x-ray mirror and a two-bounce Ge(220) crys-
TABLE I. A summary of the growth conditions for the GaN:Mg layers studied in this work: growth temperature \( (T_g) \), growth pressure, V/III ratio, growth rate, \( \text{Cp}_2\text{Mg}/\text{TMGa} \) ratio, and \( \text{H}_2 \) and \( \text{N}_2 \) carrier gases flows.

| Sample | \( T_g \) \( (^\circ \text{C}) \) | pressure \( (\text{mbar}) \) | V/III ratio | growth rate \( (\mu\text{m/h}) \) | \( \text{Cp}_2\text{Mg}/\text{TMGa} \) \( (%) \) | \( \text{H}_2 \) \( (\ell/\text{min}) \) | \( \text{N}_2 \) \( (\ell/\text{min}) \) |
|--------|----------------|----------------|-------------|----------------|----------------|----------------|----------------|
| \( M_0 \) | 1120 | 100 | 906 | 0.70 | 0 | 19 | 9 |
| \( M_1 \) | 1120 | 100 | 906 | 0.70 | 0.033 | 19 | 9 |
| \( M_2 \) | 1120 | 100 | 906 | 0.60 | 0.082 | 19 | 9 |
| \( M_3 \) | 1120 | 100 | 906 | 0.70 | 0.167 | 19 | 9 |
| \( M_4 \) | 1120 | 100 | 906 | 0.70 | 0.335 | 19 | 9 |
| \( M_5 \) | 1120 | 100 | 906 | 0.60 | 0.500 | 19 | 9 |
| \( M_6 \) | 1120 | 100 | 1811 | 0.14 | 0.335 | 25 | 12 |
| \( M_7 \) | 1120 | 100 | 453 | 1.30 | 0.335 | 25 | 12 |
| \( M_8 \) | 1120 | 100 | 453 | 1.06 | 0.500 | 25 | 12 |
| \( M_9 \) | 1040 | 100 | 453 | 1.20 | 0.500 | 25 | 12 |
| \( M_{opt} \) | 1120 | 100 | 906 | 0.60 | 0.167 | 19 | 9 |

The tilt angle was determined by the Williamson-Hall plots of the symmetric 0002, 0004, and 0006 diffraction peaks of GaN, and the magnitude of the Burger vector along the \( c \)-axis \( (b_c = 0.5185 \text{ nm}) \) was used for an estimation of the screw-type dislocation density. For the edge dislocation density estimation, a method proposed by Srikant \textit{et al.}, \cite{23} where the full-width at half-maximum (FWHM) of the rocking curves from the asymmetric 10\(\text{I}1 \), 10\(\text{I}2 \), 10\(\text{T}3 \), 10\(\text{T}4 \) 10\(\text{T}5 \), and 3052 diffraction peaks and the Burger vector along the \( a \)-axis \( (b_a = 0.3189 \text{ nm}) \) are used.

The structural quality of selected samples was further assessed by transmission electron microscopy (TEM) using the double corrected Linköping FEI Titan\(^3\) 60-300 microscope, operated in scanning TEM (STEM) mode and 300 kV. Images were acquired using annular dark-field (ADF) and annular bright-field (ABF) detectors (collection angles \( \sim 21 - 200 \) and \( \sim 4 - 43 \text{ mrad} \) respectively), revealing both atomic number and strain contrast. Electron energy loss spectroscopy (EELS) thickness measurements were performed by acquisition using a Gatan GIF Quantum detector with 10 ms exposure time, 0.25 eV/channel dispersion \( (-50.0 \text{ eV to } 462.0 \text{ eV}) \), and convergence and collection angles of 22.0 and 15.0 mrad respectively. Calculations were done in the embedded function in Gatan Digital Micrograph software. Samples were mechanically cut and mounted in Ti grids, followed by mechanical thinning and Ar-ion milling (Gatan PIPS Model 691). This produced two projection directions: \([1100]\) and \([1120]\).

The free-hole concentration and mobility of the as-grown and annealed samples were determined by Hall-effect measurements performed at room temperature in Van der Pauw configuration using a Linseis HCS1 instrument. For this purpose, Ni/Au \( (5 \text{ nm}/250 \text{ nm}) \) ohmic contacts were deposited by thermal evaporation and annealed at 450 \( ^\circ \text{C} \) in air. Note that the Hall-effect measurements of as-grown and annealed samples were performed on the same piece for a given growth condition. The effective dopant concentration \( N_A - N_D \) in samples \( M_1 - M_9 \) was extracted by capacitance-voltage C-V measurements, using a Hg-probe setup with a 4284A LCR meter from Agilent. The C-V data were acquired in series measurement mode in the 1 - 10 kHz frequency range.

The concentrations of Mg dopants, H and C impurity, the root mean square surface roughness, the density of screw and edge type dislocations and the free-hole concentration in as-grown GaN and the free hole concentration, mobility and resistivity of the respective thermally activated GaN samples are summarized in Table II.

III. RESULTS AND DISCUSSION

A. Mg incorporation

Figure H shows the Mg and H concentrations in the GaN:Mg layers as a function of the ratio between the dopant precursor and the TMGa in the gas phase, \( \text{Cp}_2\text{Mg}/\text{TMGa} \). As expected, \( \text{Cp}_2\text{Mg}/\text{TMGa} \) has a significant effect on the Mg incorporation. For samples \( M_1 - M_5 \) grown under optimized growth conditions of V/III ratio of 906 and growth rate of 0.60-0.70 \( \mu\text{m/h} \), [Mg] increases linearly with \( \text{Cp}_2\text{Mg}/\text{TMGa} \). A maximum Mg
TABLE II. Concentrations of Mg dopants ([Mg]), H ([H]) and C ([C]) unintentional impurities, root mean square (RMS) surface roughness, screw (Nₛ) and edge (Nₑ) dislocation densities, hole concentration (pₒ) in the as-grown GaN:Mg layers, and hole concentration (p), mobility (μ), and resistivity (ρ) in the respective layers after annealing.

| Sample | [Mg] (cm⁻³) | [H] (cm⁻³) | [C] (cm⁻³) | RMS (nm) | Nₛ (cm⁻²) | Nₑ (10¹⁶cm⁻³) | pₒ (10¹⁷cm⁻³) | p (cm³/V.s) | ρ (Ωcm) |
|--------|-------------|------------|------------|---------|-----------|--------------|--------------|-----------|---------|
| M₀     | 0           | 1.5 x 10¹⁸ | 9.9 x 10¹⁵ | 0.15    | 1.0 × 10⁷ | 5.0 × 10⁶   | -            | -         | -       |
| M₁     | 2.5 x 10¹⁸  | 2.4 x 10¹⁸ | 9.8 x 10¹⁵ | 0.22    | 3.8 x 10⁷ | 5.9 x 10⁸   | N/A          | N/A       | -       |
| M₂     | 6.0 x 10¹⁸  | 4.8 x 10¹⁸ | 6.0 x 10¹⁵ | 0.22    | 4.0 x 10⁷ | 6.2 x 10⁸   | 8.4          | 5.8       | 1.08    |
| M₃     | 1.6 x 10¹⁹  | 1.5 x 10¹⁹ | 1.5 x 10¹⁶ | 0.19    | 2.1 x 10⁷ | 5.4 x 10⁸   | 0.76         | 6.5       | 1.08    |
| M₄     | 2.4 x 10¹⁹  | 2.2 x 10¹⁹ | 1.5 x 10¹⁶ | 0.19    | 3.7 x 10⁷ | 4.8 x 10⁸   | N/A          | 5.3       | 1.30    |
| M₅     | 6.1 x 10¹⁹  | 9.6 x 10¹⁸ | 8 x 10¹⁵  | 0.26    | 2.9 x 10⁷ | 8.1 x 10⁸   | 9.6          | 2.3       | 2.90    |
| M₆     | 1.6 x 10¹⁹  | 1.3 x 10¹⁹ | 5.3 x 10¹⁵ | 0.17    | 3.0 x 10⁷ | 7.4 x 10⁸   | 3.1          | 8.9       | 0.88    |
| M₇     | 7.0 x 10¹⁹  | 1.4 x 10¹⁹ | 2.1 x 10¹⁵ | 0.20    | 2.4 x 10⁷ | 9.0 x 10⁸   | 0.61         | 0.9       | 7.78    |
| M₈     | 6.7 x 10¹⁹  | 1.4 x 10¹⁹ | 2.3 x 10¹⁶ | 0.45    | 5.4 x 10⁷ | 7.0 x 10⁸   | 0.02         | 1.1       | 8.73    |
| M₉     | 1.1 x 10²⁰  | 2.2 x 10¹⁹ | 3.1 x 10¹⁷ | 1.00    | -         | -            | N/A          | N/A       | -       |
| M₉₉₉₉ | 1.6 x 10¹⁹  | 1.5 x 10¹⁹ | 1.5 x 10¹⁶ | 0.18    | 1.6 x 10⁷ | 4.3 x 10⁸   | 1.67         | 8.4       | 10      | 0.77    |

The concentration of ~ 6.0 x 10¹⁹ cm⁻³ at Cp₂Mg/TMGa = 0.5% is reached. We deduce a dopant incorporation efficiency of 0.24 from the dependence of the atomic fraction of magnesium [Mg]/[Ga] in the lattice as a function of the Cp₂/TMGa flux ratio in the vapour phase (see Fig. S1 in the supplementary material). The observed trends (Fig. 1 and Fig. S1) are consistent with previously reported results for MOCVD GaN:Mg layers. [2, 5, 26, 27]

In order to explore the limitations in Mg incorporation levels, additional GaN:Mg layers, M₆ - M₉, with the two highest Cp₂Mg/TMGa ratios of 0.335% and 0.5% but with different V/III ratios, temperatures and growth rates were prepared. Samples M₆ and M₇ were grown with the same Cp₂Mg/TMGa ratio of 0.335% as for sample M₄ but with twice as high V/III ratio of 1811 and a reduced by half V/III ratio of 453 , respectively. It can be noted that for constant Cp₂Mg/TMGa ratio the increase of the V/III ratio leads to 1.5 times lower Mg concentration, bringing it below the threshold for PID formation (for details on PID, see Sec. III B). When the V/III is decreased to 453, a significant increase in [Mg] to 7.0 x 10¹⁹ cm⁻³ is observed (M₇). This is almost three times higher compared to the respective [Mg] in M₄ with a V/III ratio of 906. Note, that higher gas carrier flows of H₂ = 25 ℓ/min and N₂ = 12 ℓ/min (Table I) were used for M₇ but their ratio was kept the same as for the case of samples M₀ - M₄. A detailed study on the effects of carrier gas flows and composition are reported elsewhere. [28]

In a next step, a GaN:Mg with the same growth parameters as for M₇ but with a Cp₂Mg/TMGa of 0.5% was grown (sample M₈) to investigate whether Mg incorporation can be further boosted by increasing the Cp₂Mg/TMGa at these specific growth conditions. Interestingly, the [Mg] in M₈ is found to be 6.7 x 10¹⁹ cm⁻³, which is very similar to the [Mg] in M₇. This indicates that at low V/III ratios a saturation of Mg incorporation is already reached at a Cp₂Mg/TMGa of 0.335%. The lower V/III ratio is associated with a higher growth rate which can shift the balance from Mg atom substitutionally replacing a Ga atom towards remaining as an adatom providing potential explanation for the observed Mg saturation. Finally, a GaN:Mg with a V/III ratio of 453, Cp₂Mg/TMGa ratio of 0.5% but at a lower temperature of 1040 °C - M₉ was grown (the rest of the samples were all grown at 1120 °C). This also led to a slightly different growth rate of 1.20 µm/h compared to M₉. The lower growth temperature resulted in high Mg incorporation level in the range of ~ 1 x 10²⁰ cm⁻³.

Within the medium-to-high Mg concentration range up to 2.4 x 10¹⁹ cm⁻³, [H] follows closely [Mg] being slightly lower than the expected 1:1 ratio (see Fig. 1 and Table I and Fig. S2). Consequently, as-grown GaN:Mg exhibits p-type conductivity even before the annealing, as will be discussed later (see Sec. III C). The H levels in all GaN:Mg layers with [Mg] above 2.4 x 10¹⁹ cm⁻³ are below ~ 3 x 10¹⁹ cm⁻³, being significantly lower than the respective [Mg] in agreement with earlier observations. [1, 2] (see Fig. 1 and Fig. S2).

Our results indicate that the Mg incorporation in hot-wall MOCVD GaN is directly related to Cp₂Mg/TMGa ratio, more specifically to the Mg precursor flow in the reactor. In addition, V/III ratio, growth rate, growth temperature and pressure are impacting the growth process and hence incorporation of impurities and formation of native defects. It has been demonstrated that for MOCVD growth the concept of supersaturation, which measures the deviation from the thermodynamic equilibrium, is suitable to evaluate the impact of growth conditions in their complexity. [29] Therefore, we present in Fig. 2 [Mg] as a function of the Ga supersaturation. The latter was calculated following Ref. [29] and the respective growth parameters in Table I. Notably, the supersaturation has a strong effect on the incorporated [Mg] for samples with constant Cp₂Mg/TMGa ratio. For instance at Cp₂Mg/TMGa of 0.335%, the decrease of supersaturation from 160 (M₄) to 79 (M₆) results in 65%
FIG. 1. Mg and H concentrations ([Mg] and [H]) determined by SIMS as a function of \( \text{Cp}_2 \text{Mg} / \text{TMGa} \) ratio in the as-grown GaN:Mg layers. Squares and triangles represent [Mg] and [H], respectively. The sample labels according to Table I are indicated in the figure. The blue line represents a linear fit to the [Mg] data points for samples \( M_1 \) - \( M_5 \) grown under the same growth conditions.

The decrease of [Mg] while increasing it to 184 (M7) leads to a steep rise of [Mg] by 165%. When the Ga supersaturation for constant \( \text{Cp}_2 \text{Mg} / \text{TMGa} \) of 0.5% increased from 184 (M4, M6 and M7), the increase of Ga supersaturation was achieved by varying the TMGa flow, and by decreasing the growth temperature for the latter case (M8 - M9). The ratio \( F \) (the ratio of the input mole fraction of H2 to the total amount of the two carrier gases, H2 and N2), which also strongly affects the supersaturation is the same for all samples. These observations show that the Mg incorporation in GaN is a multi-dimensional process and the trends related to different growth parameters e. g. V/III ratio, growth temperature, carrier gas composition etc., can be assessed by taking into account only the \( \text{Cp}_2 \text{Mg} \) precursor flow and the Ga supersaturation. Clearly, Mg incorporation follows the same trend as Ga supersaturation i. e. the available Ga species in the reaction. Since Mg atoms compete with Ga atoms for incorporation at the same lattice site, at a constant Ga supersaturation the balance of Mg/Ga atoms incorporation is shifted more towards the Mg side when the \( \text{Cp}_2 \text{Mg} \) precursor flow i. e. the available Mg atoms in the reaction are increased.

O and Si impurity levels in our GaN:Mg layers were found to be at the SIMS detection limit of \((3-5) \times 10^{15} \text{ cm}^{-3}\) in all samples. Hence only C from the unintentional impurities is further discussed in its potential role as a compensating donor and a trap affecting the free hole properties. The respective C concentrations in the GaN:Mg films are presented in Fig. 3 as a function of Ga supersaturation. An overall linear increase in [C] with increasing Ga supersaturation is evident independently of whether the change in supersaturation is achieved via varying V/III ratio or growth temperature. The increase in [C] at constant supersaturation could be associated with the increase of the \( \text{Cp}_2 \text{Mg} / \text{TMGa} \) ratio. \[30\]

B. Effect of Mg on the extended defects and surface morphology

As can be seen from Table II, a small variation of the screw-type dislocations density upon increasing the Mg concentration is observed in the as-grown GaN:Mg layers. The \( N_S \) remains very similar to the value found in the undoped GaN layer. On the other hand, \( N_E \) increases from \((5-6) \times 10^8 \text{ cm}^{-2}\) to \((7-9) \times 10^8 \text{ cm}^{-2}\) for [Mg] \( \geq 2.4 \times 10^{19} \text{ cm}^{-3}\). For a comparison, in Mg doped InN layers, the edge dislocation density remains similar with increasing Mg, while screw dislocation density increases accompanied by a polarity inversion. \[31\]

STEM analysis was performed to provide further insight into the microstructure of the GaN:Mg layers. The results shown in Fig. 4 verify that extended defects are
FIG. 3. Carbon concentration ([C]) in the as-grown GaN:Mg layers vs Ga supersaturation. The sample labels according to Table I are indicated in the figure. The same as in Fig. 1, blue symbol indicate samples grown with 19 ℓ of H$_2$ and 9 ℓ N$_2$ while green symbols indicate samples grown with 25 ℓ of H$_2$ and 12 ℓ N$_2$.

not generated in layers doped with Mg up to 1.6 × 10$^{19}$ cm$^{-3}$ (Fig. 4 (a) and (b)). For [Mg] = 2.4 × 10$^{19}$ cm$^{-3}$ (sample M$_4$) sporadic PID defects are present (Fig. 4 (c) and (e)). Incorporating more Mg (sample M$_8$ with [Mg] = 6.7 × 10$^{19}$) leads to a notable increase in PID density (Fig. 4 (d)). The average width of the PIDs in the latter case is estimated to be 5.4 ± 0.9 nm (Fig. 4 (d)), in range with previously reported values. [2, 3] Polarity inversion inside the pyramids is confirmed by annular bright-field (ABF) STEM (see Fig. S3 in the supplementary material). STEM analysis further revealed segregation of Mg at the PID (0001) facet (Fig. S3) in concordance with earlier observations. [2, 3] Additionally, our comprehensive EELS analysis reveals that Mg atoms are segregated not only at the (0001) planes of the PIDs, but they are also found at their inclined facets (Further details on the EELS analysis will be reported elsewhere).

A crude estimate of the PID density results in a range between 1.9 and 8.9 × 10$^{16}$ cm$^{-3}$ by sampling different regions of M$_8$ (Fig. 4 (d)). The extended range originates from a thickness measurement by EELS acquired in an adjacent region. An estimate of the number of Mg atoms associated with the PIDs can be obtained following Narita et al., [2, 3] for calculating the Mg atoms bound to the top facet and adding the herein observed segregation of Mg to the side facets. Accordingly, the [Mg] bound to the PIDs in M$_8$ is estimated to be from 1.6×10$^{19}$ cm$^{-3}$ to 7.7×10$^{19}$ cm$^{-3}$. We expect the actual value to be on the higher end of the range, since the PID density measurement was performed close to an edge where the measured thickness is on the lower end of our estimate. In comparison, the difference between [Mg] and [H] in this sample as measured by SIMS amounts to 5.3×10$^{19}$ cm$^{-3}$ (Table I and Fig. S2). It is therefore inferred that the difference between [Mg] and [H] at doping levels above 2.4×10$^{19}$ cm$^{-3}$ can be accounted for by Mg segregated at all the interfaces of the PIDs.

The presence of PIDs is also expected to affect the surface morphology, in particular for the high-defect-density layers with [Mg] ≥ 2.4 × 10$^{19}$ cm$^{-3}$. The UID GaN and GaN:Mg layers M$_0$ - M$_5$ grown with the optimal growth conditions exhibit step-flow growth mode and the terrace size is similar for the undoped and doped layers up to [Mg] of 6.1 × 10$^{19}$ cm$^{-3}$ as can be seen from the 5×5 µm$^2$ micrographs in Fig. 4. The root-mean-square roughness (RMS) for M$_0$ - M$_5$ is in the range of 0.15 - 0.26 nm. However, on a larger-scale, changes in morphology could be observed already at moderate Mg concentrations. Figure 6 shows representative 80×80 µm$^2$ micrographs of
selected samples. It is seen that already for Mg doping of 1.6 \times 10^{19} \text{ cm}^{-3} (M_3) there is a slight increase of RMS as compared to M_1. The RMS of sample M_4, for which sporadic PIDs start to appear, is further increased by a factor of two (Fig. 9). With further increase of Mg concentration one can observe well defined hexagonal hillocks (Fig. 4 (d)). Their density and height is increasing with increasing [Mg], which is manifested in higher RMS surface roughness. This surface deterioration can be correlated with the enhanced PID density that takes place with increasing [Mg]. Previously, it was indicated that this is a gradual process that starts already at [Mg] = 2.4 \times 10^{19} \text{ cm}^{-3} and is associated with the PID formation in the overall Ga-polar GaN. Our results indicate that this is a gradual process that starts already at [Mg] = 2.4 \times 10^{19} \text{ cm}^{-3} and is associated with the PID formation in the overall Ga-polar GaN. Note that this is not accompanied by any noticeable increase of screw dislocation densities as earlier suggested for the case of full polarity inversion in GaN and experimentally observed for InN.

For example, samples M_3 and M_8 have very similar N_S (Table II) while their PID density differ by orders of magnitude.

C. Electrical and free-hole properties

Figure II shows the net acceptor concentration N_A - N_D and the free-hole concentrations in the annealed samples. The relative difference between Mg and H concentrations with respect to the total Mg concentration, ([Mg] - [H])/[Mg] in the as-grown GaN:Mg is plotted as a function of [Mg] in Fig. 5. Assuming that all H binds to the available Mg, this fraction represents the total amount of Mg atoms not bound to H and it may include: i) Mg, acceptors not passivated by H and/or ii) Mg incorporated at a different site in the GaN crystal lattice, e. g., Mg interstitial, Mg segregated on the PID facets etc. Two distinct behaviors of (N_A - N_D)/p and ([Mg] - [H])/[Mg] can be discerned below and above [Mg] = 2.4 \times 10^{19} \text{ cm}^{-3}

Up to 2.4 \times 10^{19} \text{ cm}^{-3} the N_A - N_D follows closely the [Mg] indicating that Mg is incorporated as an acceptor within this doping range. A certain fraction between 4% and 21% of [Mg] remains non-passivated by H (Fig. 5) and Table II. Since all Mg atoms are incorporated as acceptors (Fig. 7), this result implies that [Mg] - [H]/[Mg] represents the fraction of Mg atoms of type i), i.e. the isolated Mg acceptor not passivated by H. As a result, as-grown GaN:Mg manifests a net p-type conductivity with free hole concentration in the low to high 10^{16} \text{ cm}^{-3} range (samples M_2, M_3 and M_6), see Table II. These findings are very interesting since they are contrasted with previous reports, where [H] is equal or larger than [Mg] and indicate the expanded opportunities offered by the hot-wall MOCVD concept. The higher the non-passivated Mg fraction, the higher the free hole concentration in the as-grown GaN:Mg for this doping range. Details on exploring carrier gas composition for increasing the non-passivated Mg acceptor and free-hole concentrations will be presented elsewhere.

It is instructive to compare samples M_3 and M_6 with the same [Mg] but significantly different levels of non-passivated Mg of 1 \times 10^{18} \text{ cm}^{-3} and 3 \times 10^{18} \text{ cm}^{-3}, respectively. The comparison reveals that Mg exhibits four times higher free hole concentration before annealing with regard to M_3. Assuming
ionization energy of 180-200 meV corresponding to $N_A$ of $1 \times 10^{18}$ cm$^{-3}$ and $3 \times 10^{18}$ according to Ref. [7], the maximum free-hole concentrations in the as-grown $M_3$ and $M_6$ layers are expected to be in the mid and high $10^{16}$ cm$^{-3}$ range, respectively. The $p_0$ values measured for $M_3$ and $M_6$ are lower indicating certain degree of compensation. We recall that $M_6$ is grown at higher V/III ratio as compared to $M_3$, i.e., at lower supersaturation (Table II and Fig. 3). Under the N-rich conditions, typically employed in MOCVD growth, the $C_Ga$ acting as a donor has the lowest formation energy in $p$-type GaN. [12] Although $[C]$ is at least two orders of magnitude lower than the non-passivated isolated Mg acceptors ($[Mg]$ - $[H]$), it is close to the observed free-hole concentrations in the as-grown layers and will interfere negatively with the $p$-type conductivity. This can explain the observed lower free-hole concentration before annealing in $M_3$ as compared to $M_6$. After annealing the free-hole concentration in $M_6$ is also higher as compared with $M_3$, correlating with $N_A - N_D$. In this case, $N_A - N_D$ is dominated by the thermally activated Mg acceptors resulting from the dissociation of H from the Mg-H complexes in the as-grown samples.

The increase of Mg concentration above 2.4 $\times 10^{19}$ cm$^{-3}$ leads to a decrease of $N_A - N_D$ which becomes significantly lower than $[Mg]$ (Fig. 7), consistent with previous reports. [2, 8] The C-V measurements show that 72% to 92% of the incorporated Mg atoms are not electrically active. This doping range corresponds to $[H] \ll [Mg]$ in the as-grown samples as can be seen from Figs. 1 and 2 and the respective fraction of Mg not-bound to H is in the range of 74% to 84% with no distinct dependence on $[Mg]$ (Fig. 5). These results imply that the number of Mg atoms not binding to H in the as-grown layers could account to a large extent for the difference between the net acceptor and Mg concentrations in the respective samples after annealing. In other words, the fraction $([Mg] - [H])/[Mg]$ corresponds to scenario ii) in which Mg incorporates at non-acceptor sites in the GaN crystal lattice.

This is reflected in a significant reduction of free-hole concentration in the annealed samples (Fig. 7). Note that the lower $N_A - N_D$ (Fig. 7) the lower the free hole concentration in the as-grown (Table II and Fig. 3) and annealed (Fig. 7) samples. Our STEM analysis shows that the observed large fraction of electrically inactive Mg could potentially be explained by the estimated concentration of Mg segregated at the PIDs. However, the fraction of Mg not bound to H is very similar for samples $M_5$ and $M_6$ (Fig. 8) while their $N_A - N_D$ differs significantly (Fig. 7). For example, sample $M_5$ with inactive Mg fraction of 84% has more than twice as high net acceptor concentration than $M_5$ with 74% of inactive $[Mg]$ non-bound to H, while the opposite could be expected. In fact, in $M_5$, the concentration of Mg-H complexes before annealing is $9.6 \times 10^{18}$ cm$^{-3}$ (assuming all available H binds to Mg), which compares well with the $N_A - N_D$ of $1.7 \times 10^{19}$ cm$^{-3}$ after annealing. This suggests that no significant concentrations of compensating donors, most notably the $V_N$ and its complexes with Mg, are generated. In samples $M_7$ and $M_8$, the Mg-H levels before annealing are slightly higher as compared to $M_5$, while $N_A - N_D$ are significantly lower, i.e. $(5-8) \times 10^{18}$ cm$^{-3}$, indicating that compensating defects are present in large densities. Both $M_7$ and $M_8$ are grown at lower V/III ratio, which can explain an enhanced formation of $V_N$ in this case. Our findings strongly suggest that compensating donors, likely $V_N$ and its complexes with Mg are generated for highly Mg doped GaN grown at higher su-

![FIG. 7. Net acceptor concentration $N_A - N_D$ (from C-V measurements) and free-hole concentrations (from Hall) in the annealed $p$-GaN as a function of $[Mg]$. The red and orange curves are guide to the eye. The blue solid line corresponds to $N_A - N_D = [Mg]$.](Image)

![FIG. 8. Fraction of non-passivated Mg, $([Mg] - [H])/[Mg]$ in the as-grown GaN:Mg layers as a function of Mg doping.](Image)
persaturation and which play a significant role for the observed reduction of free-hole concentration (Fig. 7). The increase of [Mg] above $1 \times 10^{20}$, does not alter the fraction of non-passivated electrically inactive Mg. In this case, however, the high [C] levels of $3.1 \times 10^{17}$ cm$^{-3}$, due to the significantly lower growth temperature, lead to additional compensation and the GaN layer shows semi-insulating behavior.

It has been shown that by growing GaN:Mg on GaN substrates, with significantly lower density of dislocations, the free hole concentration in the homoepitaxial layers can be significantly enhanced. To reduce the dislocation density in our heteroepitaxial case we grew $M_{\text{opt}}$ consisting of a 550-nm-thick GaN:Mg layer on 1-μm-thick undoped GaN buffer layer using the same growth conditions as for sample $M_3$. Both screw- and edge-type dislocation densities of $M_{\text{opt}}$ show the lowest values of $1.6 \times 10^7$ cm$^{-2}$ and $4.3 \times 10^8$ cm$^{-2}$, respectively, among all samples (Table II). $N_S$ and $N_E$ in the GaN:Mg are likely to be even lower since the measured dislocation densities are averaged over the entire layer thickness. The free-hole concentration in the as-grown $M_{\text{opt}}$ is increased more than twice as compared to the respective value in $M_3$. If similar approach is employed for lower Mg doping levels, e. g. such as in $M_2$, free-hole concentrations in the low to mid $10^{17}$ cm$^{-3}$ range can be potentially achieved without the need of thermal activation. The free-hole concentration of $8.4 \times 10^{17}$ cm$^{-3}$ and the resistivity of 0.77 Ω.cm in the annealed sample are among the best results reported in literature. These results demonstrate that the hot-wall MOCVD capabilities can be expanded to deliver state-of-the-art p-type GaN enabling development of power diodes and vertical transistors, and normally-off HEMTs, for example.

IV. CONCLUSIONS

We have explored hot-wall MOCVD for the growth of Mg doped GaN. A detailed study of growth and doping conditions impacts on the incorporation of Mg, H and C is presented and the findings are discussed in terms of Ga supersaturation. Our results clearly indicate that Ga supersaturation is a universal parameter that can be conveniently used for the optimization of Mg incorporation and C reduction in MOCVD of GaN instead of multiple growth parameters independently used for the same purpose.

In the low doping range ([Mg] $\leq 2.4 \times 10^{19}$ cm$^{-3}$), a large fraction (up to 21%) of active Mg acceptors (not passivated by H) is evidenced in as-grown material. Hence, hot-wall MOCVD provides an intriguing opportunity to exploit the technique for delivering lightly p-type doped material for e. g. vertical MOSFETs, without the need of post-growth annealing. In the annealed samples, $N_A - N_D$ is found to closely follow [Mg] indicating that all Mg atoms are incorporated as acceptors, where 96% are activated by thermal dissociation of H during post-growth annealing.

In the high doping range above $2.4 \times 10^{19}$ cm$^{-3}$, increased Mg leads to the generation of PIDs. It is shown that this can be correlated with the formation of hexagonal hillocks, the density and height of which increase with increasing [Mg]. This gradual surface morphology deterioration can be clearly observed in large-scale AFM imaging while on a smaller scale the morphology and surface roughness remain virtually unchanged. STEM analysis reveals segregation of Mg at the PIDs, sufficient to account for the observed amount of electrically inactive Mg not passivated by H. However, generation of $V_N$ and its complexes with MgGa need to be invoked to explain the considerably lower free hole and net acceptor concentrations measured in the highly doped GaN:Mg grown at high supersaturation.

Under optimized growth conditions high-quality GaN:Mg with resistivity of 0.77 Ω.cm and free-hole concentration of $8.4 \times 10^{17}$ cm$^{-3}$ corresponding to Mg acceptor activation of 5.25% has been demonstrated. These results and the established comprehensive picture of GaN:Mg growth via hot-wall MOCVD substantiate the expanded capabilities of the technique to deliver state-of-the-art p-type GaN for the development of power diodes and transistors.

SUPPLEMENTARY MATERIAL

See the supplementary material for additional details about Mg and H incorporation in the studied layers as well as on the structure of the observed pyramidal defects as revealed by STEM.

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AUTHOR DECLARATION

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available within the article and its supplementary material.

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SUPPLEMENTARY: MG-DOPING AND FREE-HOLE PROPERTIES OF HOT-WALL MOCVD GAN

Figure S1 shows how the atomic fraction of Mg in GaN lattice varies as a function of the percentage of Cp₂/TMGa precursors flux in the gas phase. Mg concentration, [Mg], is measured by SIMS and [Ga] = 4.4 × 10²² cm⁻³. The slope K of the linear fitting shows that the Mg incorporation efficiency is K ≈ 0.24 and this value is very similar to literature reported values for p-type GaN growth in cold-wall MOCVD.

FIG. S1. Mg atomic fraction in GaN as a function of Cp₂/TMGa flux ratio in the gas phase. The slope K, of the linear fit, gives the Mg incorporation efficiency.
FIG. S2. Hydrogen concentration, \([\text{Mg}]\) vs magnesium concentration, \([\text{Mg}]\). The solid line represents one to one correspondence between \([\text{Mg}]\) and \([\text{H}]\). The same notation for the symbols representing samples grown at different growth conditions as in Fig. 1 is used.

FIG. S3. Polarity inversion inside the pyramids. Image is acquired using annular bright-field (ABF) STEM using collection semi-angles of \(\sim 4-43\) mrad (resulting in an inverted contrast compared to HAADF, but also increased contrast of N). The layer of Mg atoms segregating at the \((0001)\) facet is marked with an arrow and the structure is shown schematically to the right. Growth direction \([0001]\) is up. Ga is shown in green and N in blue (darker green for the N-polar structure in the pyramid). The N-polar pyramid and the Ga-polar matrix are seen overlapped.