

Limitation of simple np-n tunnel junction based LEDs grown by MOVPE

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We show evidence that tunnel junctions (TJs) in GaN grown by metal-organic vapor phase epitaxy are dominated by trap assisted (Poole-Frenkel) tunneling. This stems from observations of the careful optimized doping for the TJs. Especially the p⁺⁺ and the n⁺⁺ layer are far from ideal. The n⁺⁺ layers induces 3D growth, which can be seen by a rising oxygen signal in Secondary Ions Mass Spectroscopy (SIMS). Furthermore, Mg segregation observed by SIMS indicate a depletion region of more than 10 nm. Still, we could realize TJ based LEDs with a low penalty voltage of 1.1 V and a specific differential resistance of about 10⁻² Ω.cm² at 20 mA without using an InGaN interlayer.

I. INTRODUCTION

Realization of high quality p-n homojunction based on wide bandgap semiconductors is challenging due to doping asymmetry problems [1]. GaN, with a bandgap of about 3.4 eV, does not deviate the rule. While doping n-type is rather simple, achieving decent p-type layers is not straightforward [2,3].

Intentional n-type doping of GaN is traditionally realized by substituting gallium by silicon, which allows to reach electron concentration up to 10¹⁹ cm⁻³. However, at higher doping levels, Si doped GaN layers tend to roughen or crack under tensile stress [4,5]. Recently, germanium, whose size is comparable to gallium, has been studied as alternative dopant to cover the 10¹⁹-10²⁰ cm⁻³ range [6-8]. Oxygen on N-site is an efficient n-type donor, although only few reports exist on intentional oxygen doping [9-11]. N-type doping of GaN by other chalcogens (sulfur, selenium...) substituting the nitrogen site is little investigated. However the huge size differences to nitrogen does not bode well for achieving high doping levels and the

relatively low vapor pressures of these solid easily causes strong carrier over [12,13].

For p-doped GaN, there are even less suitable acceptors. Zinc, a standard dopant in other III-V materials, is still a relatively deep acceptor in GaN (E_a~300-400 meV) [14]. Carbon exhibits an amphoteric behavior and beryllium is a shallow acceptor (E_a~100-140 meV) but suffers from severe self-compensation [15-18]. Other unsuccessful attempts were made using calcium, cadmium and mercury [19]. In the end, magnesium is the only dopant that allows a decent p-type conduction after an activation. Unfortunately, the maximum hole concentration barely reaches 10¹⁸ cm⁻³ due to the large ionization energy (E_a~150-200 meV, and thus ~0.1% of dopant ionized at RT) even though mid 10¹⁹ cm⁻³ magnesium can be incorporated before onset of compensating defects [20].

Recently, tunnel junctions (TJs) have gained interest in nitrides. Such TJs could alleviate non-equilibrium hole injection, reduced optical loss in UV-LEDs, better current spreading in higher mobility n-type layers, inter-cavity contact in

VCSEL, or simpler vertical integration of tricolor micro-LEDs for micro-displays [21-26]. Esaki junctions typically required both low or moderate bandgap materials and doping levels around 10^{20} cm^{-3} that is out of reach for GaN [27]. Nevertheless, GaN-based TJ-based LEDs reported surprisingly low penalty voltages [28-34]. The present paper aims to shed light on the efficiency of GaN based TJ grown by metal-organic vapor phase epitaxy (MOVPE). Hence, we first analyzed the crystal properties of p- and n-GaN layers separately. Then, we use them in simple np-n structure and corresponding TJ-based LEDs. Based on those findings we discuss the tunneling mechanism.

II. EXPERIMENTAL DETAILS

All the samples were prepared in a 3x2" EpiQuest vertical showerhead MOVPE reactor. For Si and Mg doping pure H_2 was used as carrier gas. Both Mg:GaN and Si:GaN layers were grown using a V/III ratio of 7200, at a pressure of 150 Torr and at temperatures ranging from 975 to 1100°C with a growth rate of 500 nm/h. Bisethylcyclopentadienyl-Mg (EtCp₂Mg), tetramethyl-silane (TMSi), trimethyl-gallium (TMG) and ammonia (NH_3) were used as metalorganic precursors. For the growth of the QWs, the carrier gas was switched to pure N_2 and triethyl-gallium (TEGa) and trimethyl-indium (TMIn) were used as precursors.

Electrical characterization was performed by Hall effect measurements in Van-der-Pauw geometry. For Si doping levels below 10^{19} cm^{-3} Hall effect measurements and secondary ion mass spectroscopy (SIMS) concentrations agreed. Above that limit, the dopant concentration was measured by SIMS or given as extrapolation of our calibration. Photoluminescence (PL) was performed at room temperature using a He-Cd laser ($\lambda_{\text{ex}}=325 \text{ nm}$, $P=1 \text{ mW}$). For Mg:GaN layer

activation, the samples were annealed at 700°C for 5 minutes under N_2 . Surface morphologies were investigated by a Nanocute Hitachi atomic force microscope (AFM) working in tapping mode. Scanning transmission electron microscopy (STEM) pictures were recorded using a SU-70 Hitachi microscope with an operating voltage of 5 kV.

TJ based LEDs were processed by standard photolithography. L-shape mesa of 0.0675 mm^2 were prepared using Cl_2 dry etching. Then, the samples were annealed for 30 min at 750°C in N_2 ambient. Finally, Ti/Al electrodes were simultaneously deposited by e-beam lithography on both n-GaN terminals and annealed 5 min at 625°C in pure N_2 . For the reference LED, p-GaN top layer was contacted by a Ni/Au electrode annealed 10 min in air at 500°C.

III. RESULTS AND DISCUSSION

Fig. 1 summarizes the morphological, optical and electrical properties of 500 nm thick Mg doped GaN layers grown at various Mg/Ga ratios, which reflects current literature. The near band edge (NBE) emission of the PL slowly decreases and a characteristic donor-acceptor pair (DAP) related signals appear while increasing the Mg content [35]. A strong blue-band (BB) dominates the PL spectrum when the hole concentration reaches its maximum around few 10^{17} cm^{-3} at RT, corresponding to a Mg concentration of few 10^{19} cm^{-3} [36]. Further increase of the Mg content leads to the apparition of various compensating defects and pyramidal inversion domains (IDs) [37]. Simultaneously, the doping level drastically decreases and the p-GaN layers become resistive. At a Mg concentration of a few 10^{20} cm^{-3} - which is the doping level typically required for TJs - the surface is rough and decorated with large hexagonal hillocks from N-polar inversion domains (IDs) likely induced by the formation of

Mg₃N₂ sub-structures [38,39]. A high density of defects is indicated by the absence of any PL signal.

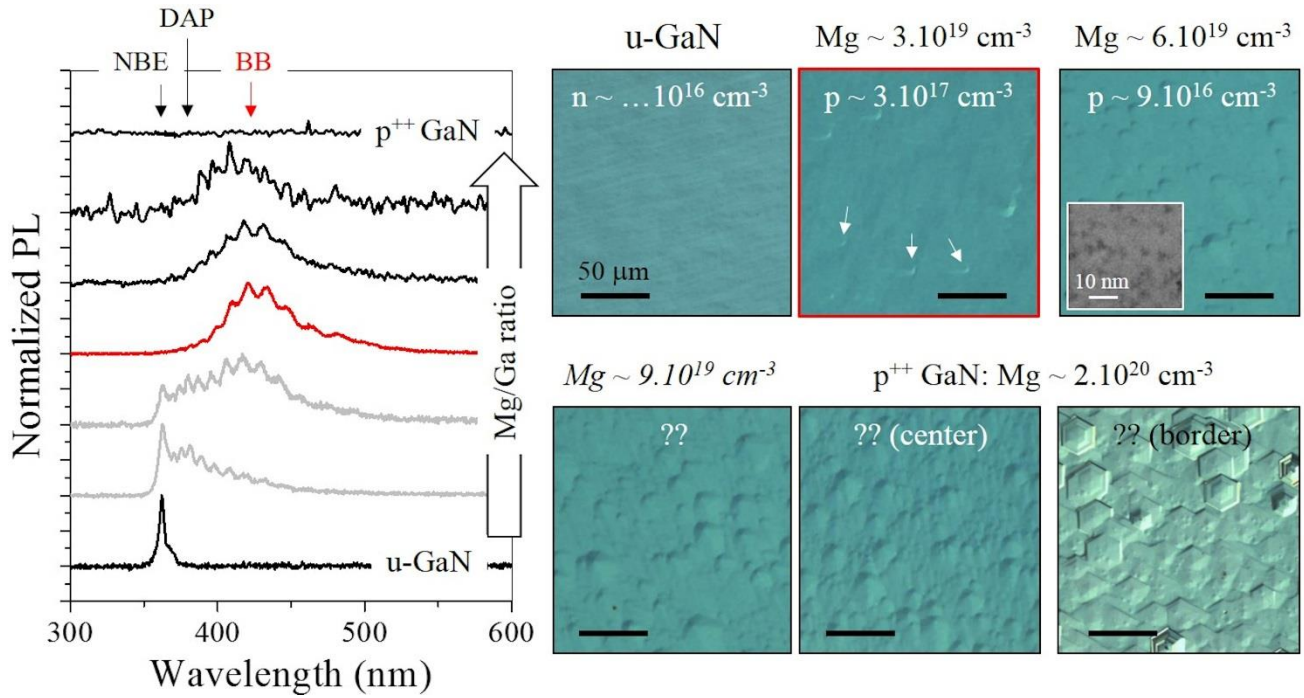


FIG. 1. Normalized PL emission spectra and optical microscope pictures of post-annealed GaN grown at 975°C at various Mg/Ga ratio. The inset shows the pyramidal IDs observed in cross-section by TEM.

Fig. 2 shows a similar study on 500 nm thick (crack-free) Si doped GaN layers grown at various Si/Ga ratio using TMSi. The electron concentration increases with the Si/Ga ratio both for growth temperature of 975 and 1100°C. At Si concentration exceeding $1.5 \times 10^{19} \text{ cm}^{-3}$, the surface show large scale-like islands of different height separated by pitted boundaries. Further increase of the Si level leads to extremely rough surface characterized by larger pits and smaller grains, similar to previous reports [5,40]. Under typical

MOVPE growth conditions and for a concentration higher than 10^{19} cm^{-3} , Si acts as an anti-surfactant. It was suspected to locally form Si-Ga-N substructures, which initiate a SiN_x nano-masking that promotes 3D growth [41,42]. However, based on fine HR-TEM analyses it is more likely a SiGa₃ monolayer that inhibits the 2D growth [43]. Thus at the doping levels used for TJ application, i.e. above 10^{20} cm^{-3} , Si behaves as an anti-surfactant rather than a dopant, as it is incorporated inactively in SiGa₃.

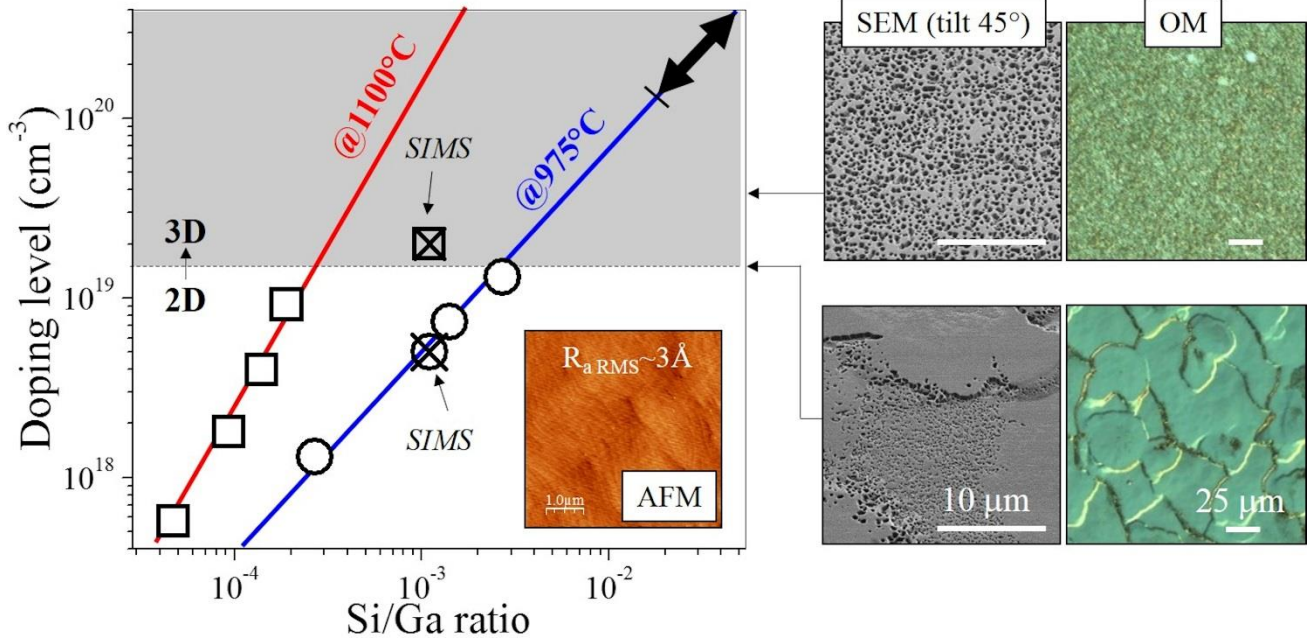


FIG. 2. Doping levels and surface morphology of Si:GaN layers grown at different Si/Ga ratio. A transition from 2D toward 3D growth occurs for Si concentration higher than 10^{19} cm^{-3} . The thick left-right arrow indicates the conditions typically required for the growth of TJ.

We observed that the 2D-3D transition always happens around a doping level of 10^{19} cm^{-3} whatever the growth temperature. Thus the 2D-3D transition is driven by the actual Si concentration on the surface and not due to surface kinetics of Ga or the precursor we used. The different Si/Ga ratio is due to the decomposition kinetics of the TMSi, whose decomposition temperature is estimated to be around 1000°C [44]. In other words, the supply of active Si, i.e. the effective Si/Ga ratio in the gas phase, depends with TMSi on the growth temperature. SiH_4 , a more common precursor, does not present such disadvantage since it is fully decomposed around 700°C , and thus usually no temperature dependence of the roughening is reported.

Fig. 3 shows the SIMS profiles of two Si and Mg doped GaN layers grown successively at four different temperatures without varying the other growth parameters, i.e. keeping the NH_3 , TMGa, TMSi and EtCp_2Mg flows constant. Unlike TMSi, the EtCp_2Mg decomposition and further Mg incorporation is stable over temperatures ranging

from 975 to 1100°C . Mg piled-up at the surfaces during the 5 min growth interruptions (GI) used to ramp up and stabilize the temperature between each step, which indicates we doped below the absolute solubility limit. Both for Si:GaN and Mg:GaN, the carbon level increases from 10^{16} to 10^{17} cm^{-3} when the growth temperature decreases from 1100 to 975°C . This is commonly attributed to the lower decomposition rate of NH_3 and less active H readily available to react and etch away hydrocarbons species present on the surface and is also observed at similar level in undoped layers. Such levels does not significantly affect the GaN conductivity. Interestingly, the O signal is very different for both layers. While the background level remains below 10^{16} cm^{-3} throughout the p-GaN growth, the O concentration increases of one order of magnitude when the n-GaN doping level reaches about 10^{19} cm^{-3} . Conversely, once the Si level decreases below 10^{19} cm^{-3} , the O concentration slowly returns to the background level. Since 3D growth starts above 10^{19} cm^{-3} as mentioned previously, and 3D growth generates

additional semi- and non-polar facets which are usually prone to higher O incorporation [45], the O signal correlates with the 2D-3D growth transition.

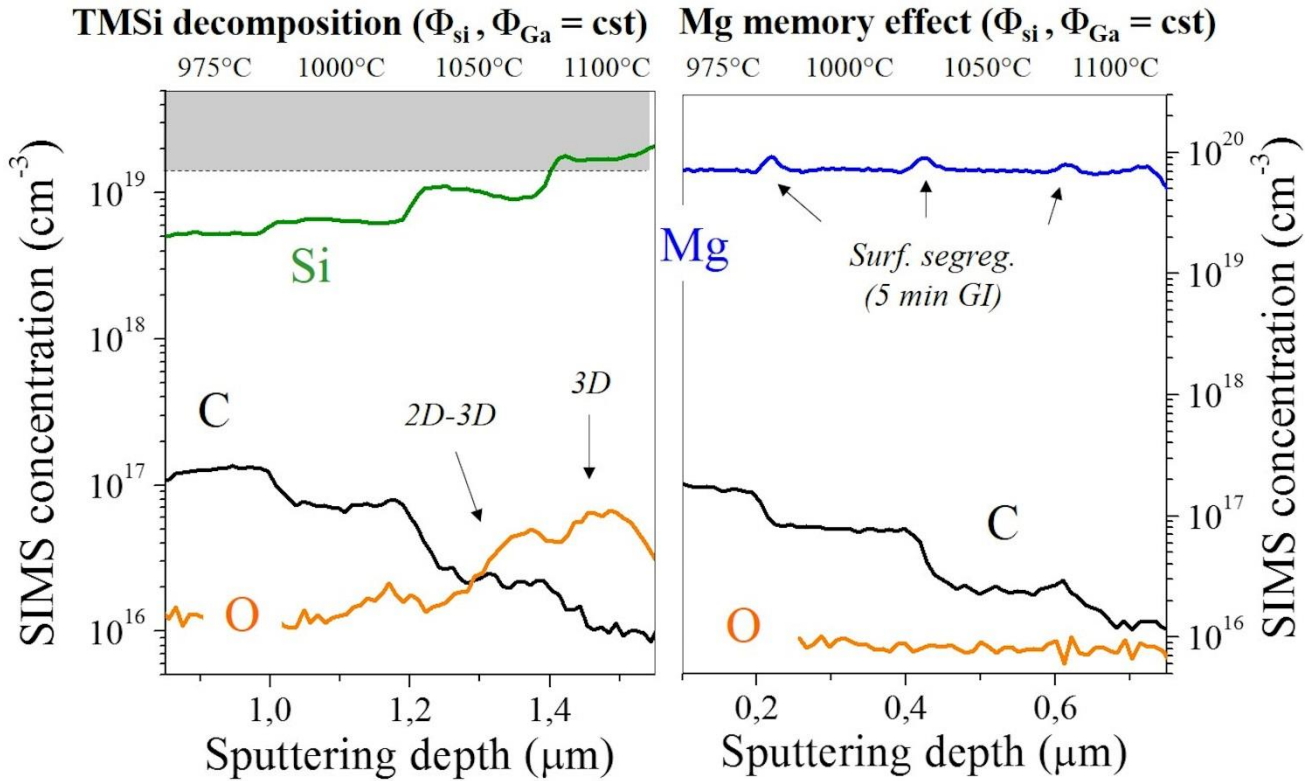


FIG. 3. SIMS analysis of n-GaN (left) and p-GaN (right) samples grown at different temperatures. The O signal increases of one order of magnitude when the Si level is higher than $10^{19}/\text{cm}^3$, i.e. when the growth mode changes from 2D to 3D. The Mg incorporation is constant over the full temperature range between 975 and 1100°C but Mg tends to accumulate on the surface during each growth interruption.

Fig. 4 presents two additional evidences for correlation of the O level with 3D growth, triggered with and without SiN_x nanomasking. For the first sample, 600 nm n-GaN is grown at 975°C with a linear increase of the Si concentration up to several 10^{19} cm^{-3} , followed by a 600 nm u-GaN to recover a smooth surface. After a 15 min growth interruption, the same sequence is repeated at 1100°C. It is worth noting that the O concentration increases as soon as the 3D growth is triggered by the Si nanomasking (black dots and arrows on Fig. 4 (left)). Conversely, the O level slowly decreases upon u-GaN which again smooths the surface. The

second sample shows a similar behavior (Fig. 2 (right)) during a u-GaN template growth on sapphire. Here intentionally, 3D islands are obtained by annealing a nucleation layer deposited at low temperature (LT). At this point, the O level is around 10^{18} cm^{-3} . Further regrowth of the 3D nuclei at high temperature (HT) smooths the surface. Since this means that the semipolar facettes are vanishing, the O concentration also decreases until it reaches its background level of about 10^{16} cm^{-3} for O incorporation on a flat (0001) surface.

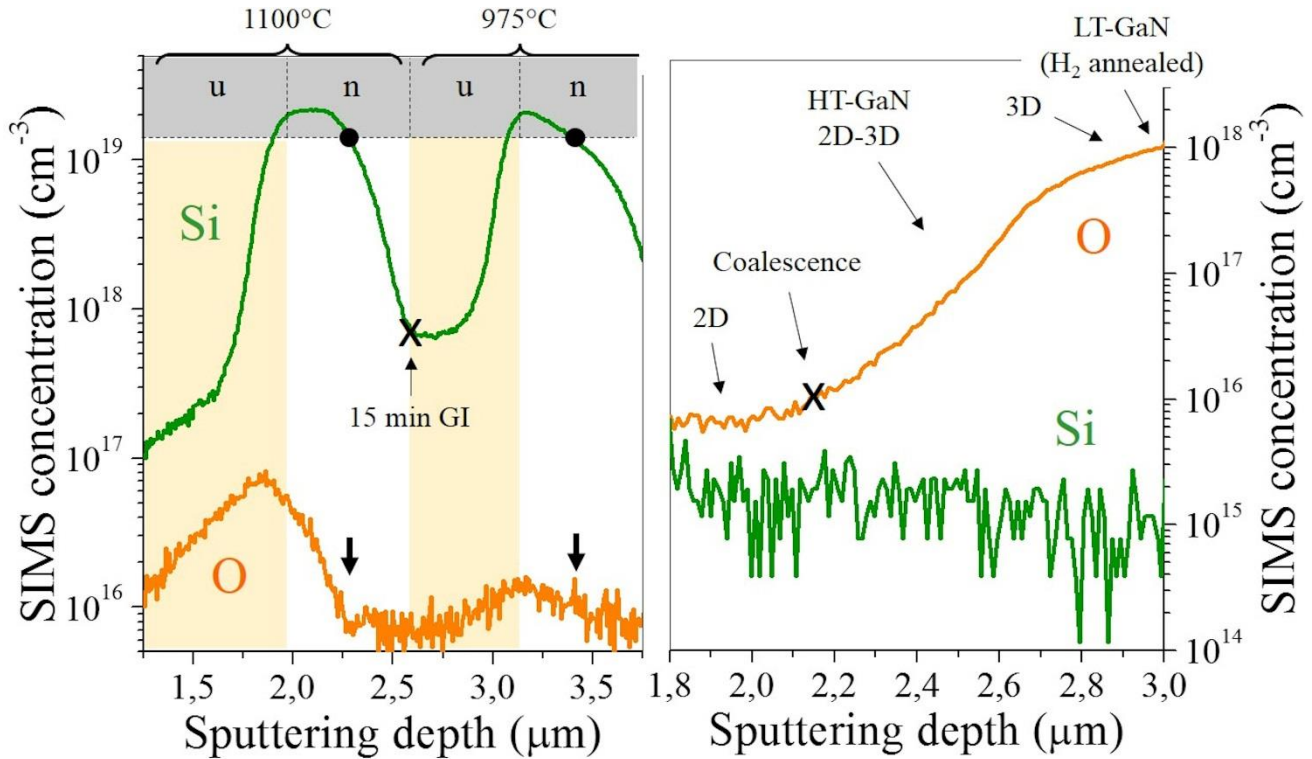


FIG. 4. SIMS analysis of n-GaN and u-GaN layers. An increase of the O level is observed each time the 3D growth mode is triggered, either by a Si concentration higher than 10^{19} cm^{-3} (left) or a H_2 annealed LT buffer layer (right). Further GaN overgrowth of the rough surface with proper conditions leads to coalescence and the O level simultaneously decreases to below 10^{16} cm^{-3} .

Fig. 2 and 3 indicate a rather low doping limit on both Si:GaN and Mg:GaN, which is theoretically unsuitable for a TJ. However, driven by many successful reports found in the literature, we compared the SIMS profile of an np-n LED (no n^{++}/p^{++} layers at the p-n junction) and a TJ based LED. Fig. 5 (left) shows the results of the np-n LED structure featuring the highest doping level as determined from Fig. 1 and 2 ($\sim 10^{19}$ and few 10^{19} cm^{-3} for Si and Mg respectively). The O level exhibits a peak around the QWs position, most likely due to the residual O found in the N_2 carrier gas required for the InGaN growth (n- and p-GaN are grown under pure H_2 carrier gas, which is easier to purify and present lower contamination levels). Since Mg has a strong carry over into subsequent layers [46-48], it is also incorporated in the Si:GaN top layer. Even though Mg decreases exponentially, the first ten nanometers of Si:GaN has still a higher Mg than Si concentration and hence are rather

insulating. Important to notice is that the Mg is potentially active in this region since the hydrogen level correlates with Mg, and hydrogen does not incorporate well in Si:GaN [49]. In conclusion, the electrical interface of the maximum doped np top junction is not as well defined as intended. For comparison, we realized the same structure with a 15 nm/15 nm n^{++}/p^{++} TJ inserted between the n- and p-GaN top layers. The Si and Mg levels were aimed to be around 2×10^{20} and $1 \times 10^{20} \text{ cm}^{-3}$ respectively. The SIMS analysis in Fig. 5 (right) shows that an additional O peak appears in the n^{++} region, indicating a rough interface due to 3D growth. Unfortunately, the SIMS profiles lack depth resolution, i.e. the concentration of the different elements is averaged over a larger volume, depending on the shape of the sputter crater at this position. This underestimates the concentration in these layers. Still, when reaching proper Si:GaN conditions, the rough surface can be smoothed

by a proper overgrowth. Thus, as expected from Fig. 2, the 15 nm thin n^{++} layer has a lot of nanomasking, is very rough, and likely

discontinuous, i.e. it may be even insulating in place, in short not appropriate for an abrupt TJ.

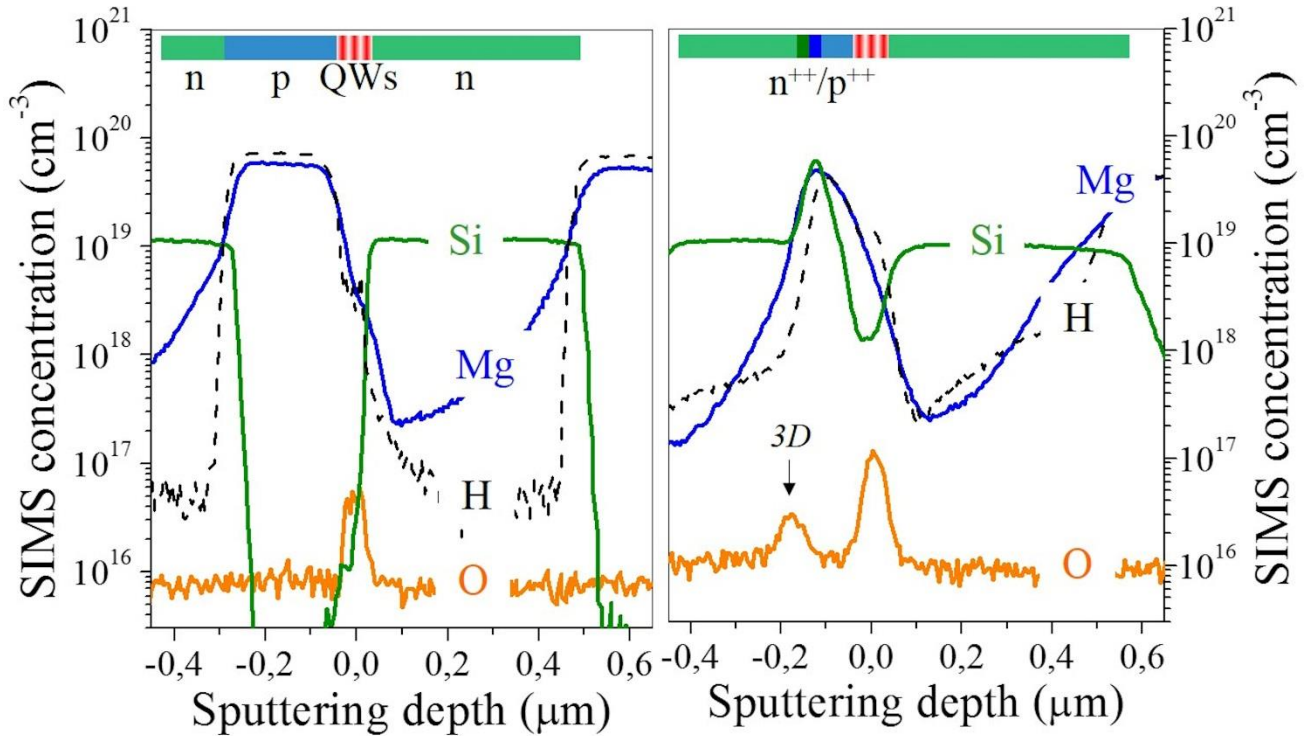


FIG. 5. SIMS analysis of the np-n (left) and TJ (right) based LEDs. Note the O peak and the Mg tail buried in the n^{++} side of the TJ. The x-axis origin has been set at the QWs position for better comparison.

Fig. 6 (left) compares the IV characteristics of three TJ based LEDs with different p^{++} and n^{++} doping levels. The reference is a standard cyan LED emitting at a wavelength of 490 nm (red curve). In good agreement with the results found in the literature, the TJ based LEDs show typical LED behavior and emit light once both Mg and Si expected levels reach about 10^{20} cm^{-3} [31]. A further increase of either dopant leads to a significantly decrease of the penalty voltage. Si seems more efficient than Mg at lowering the junction barrier, as previously observed by Kaga *et al.* [32]. Eventually, for Mg and Si levels of 10^{20} and $2 \times 10^{20} \text{ cm}^{-3}$, the TJ based LED penalty voltage is reduced to as low as 1.1 V with a specific differential resistance of about $10^{-2} \Omega \cdot \text{cm}^2$ at 20 mA. These values are surprisingly low considering the present structure does not involve any p-InGaN

interlayer that is often introduced between the p^{++} and n^{++} layers to reduce the nominal tunnel barrier height and facilitate the tunneling process [29,50].

Based on the rather poor morphological, crystalline, and electric properties of the overdoped n^{++} and p^{++} GaN layers presented in the previous sections, the results of Fig. 6 (left) seriously challenge a pure tunneling mechanism for the following reasons:

- i) The maximum free carrier density achievable in the p^{++} and n^{++} side of the junction is limited around 3×10^{19} and $2 \times 10^{19} \text{ cm}^{-3}$ respectively (assuming a complete ionization of both dopants). Therefore, the space charge region (SCR) spreads over a distance of 20 nm which is

not compatible with a Fowler-Nordheim emission, given the large bandgap of GaN.

- ii) The Mg carry over causes fairly high amount of active, i.e. not passivated, Mg ($>10^{19} \text{ cm}^{-3}$) to be incorporated in the n^{++} side. This leads to a significant compensation of the free electron density, especially at the vicinity of the interface, which widens the SCR and so further reduces the electric field across the junction.
- iii) Due to the effective doping asymmetry $p^{++} > n^{++}$ deduced from i) and ii), the SCR is shifted inside the n^{++} layer. This part of the junction is characterized by a low structural quality and a high O contamination at grain boundaries inherited from a SiN_x induced 3D growth. Worse, the presence of Mg likely induces additional defects such as IDs, or Mg-O and Mg-Si complexes. Thus, the field of the SCR is strongly affected by (various) deep centers that prevents the free carriers to directly tunnel from one side to the other one, and could even pin the Fermi level at their midgap states.
- iv) Any attempt to increase the Mg and Si concentrations up to 10^{20} cm^{-3} exacerbates

the problems discussed in points ii) and iii) even though the penalty voltage decreases.

These considerations imply that a trap-assisted tunneling, i.e. a Poole-Frenkel emission, is the main mechanism responsible for the TJs in GaN [51]. The band diagram of such TJ and the proposed tunneling mechanisms are sketched in Fig. 6 (right). Instead of a direct band to band tunneling, the carriers tunnel through defect states. Thus a less abrupt band transition can still allow relatively efficient tunneling. The tunnel current is then rather limited by the distance of the traps, and a more trap rich material could sustain higher tunnel current. It is worth pointing out a similar mechanism should occur with AlGaN. Here the problems are even worse since the larger gap would require even higher dopant concentrations and associated issues. In addition, at higher levels, the Si forms DX centers in AlGaN. Hence, overdoping would lead to more trap states. However, assuming Poole Frenkel TJ explains the recently reported relatively high conductivity of AlGaN TJ in deep UV LEDs [21].

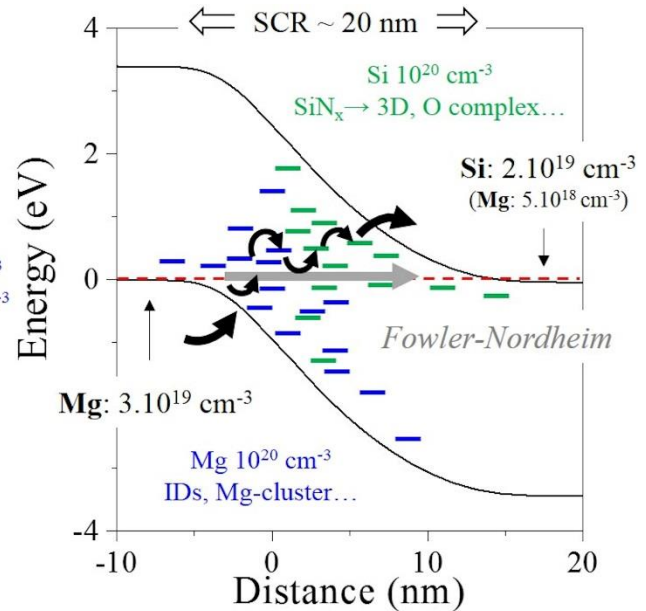
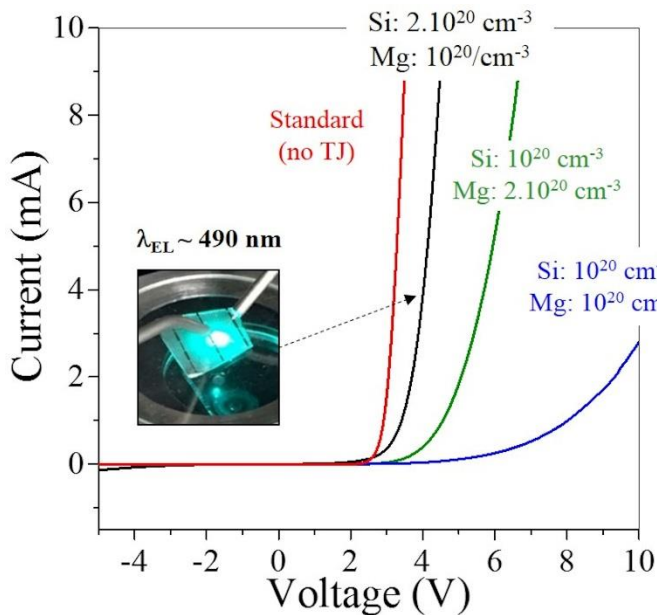


FIG. 6. I-V characteristic of standard and TJ based LEDs with different p^{++} and n^{++} doping levels (left). Band diagram of the unbiased TJ assuming that the p and n doping levels saturate at 3×10^{19} and $2 \times 10^{19} \text{ cm}^{-3}$ respectively, and taking into account the carry over of Mg (right). The curved black arrows sketches the trap-assisted tunneling (Poole-Frenkel emission). The straight grey arrow represents a pure tunneling (Fowler-Nordheim emission). Note the position of the defect levels is arbitrary.

We must stress that our results are obtained by MOVPE growth. A TJ grown by molecular beam epitaxy (MBE) does not significantly suffer from Mg carry over and Mg-H passivation, even when using NH_3 as nitrogen precursor [52]. In addition, higher doping levels for smooth layers have been achieved by MBE up to a few 10^{20} cm^{-3} [53,54].

Therefore, it seems interesting to discuss TJs found in the literature in the light of our observations and results. For instance, Young *et al.* observed the TJ formed by MBE overgrowth of a MOVPE grown p-n LED by atomic probe tomography. They observed a high O concentration at the TJ interface, most likely originating from the residual GaO formed during the sample transfer from the MOVPE reactor to the NH_3 -MBE chamber [55]. In contrast to our conclusion that O introduces midgap states in the SCR, the authors concluded the O rather establish a delta doping that strongly reduces the SCR. However, regrowth on a partly GaO covered template can easily lead to 3D growth due to different attachment rates on GaN and Ga_xO_y . More recently, Akatsuka *et al.* reported that an overlap of Mg and Si doping at the TJ interface enhances the tunneling current in MOVPE grown TJs [56]. This is counterintuitive to a sharp TJ, so the authors speculated that either GaN bandgap narrowing due to heavy doping or an advanced “2Mg+1Si” p-GaN co-doping (proposed by Katayama-Yoshida *et al.* [57]) would explain the lower resistivity of their device. However, their results are very reasonable when assuming that traps are needed to enhance tunneling. Because the strong crystal disorder introduced at the TJ interface most likely favor the creation of traps that can assist the tunneling of the carriers. A similar argument applies to the so-called polarization enhancement TJ structures.

Theoretically, the insertion of a thin InGaN interlayer between the p^{++} and n^{++} GaN allows a narrower SCR (due to the local bandgap reduction) and a higher electric field across the junction (due to built-in spontaneous and piezoelectric polarization), which is beneficial for carrier tunneling [58]. Practically, one should also consider that InGaN is grown at low temperature, and thus has a much higher defect density than GaN. In the case of MOVPE, InGaN must be grown under pure N_2 ambient and features a much higher O background (as seen in Fig. 5) while high quality n- and p-GaN requires H_2 and higher temperature. Thus, one has to choose between growing the whole TJ at same temperature under N_2 (resulting in poorer crystal quality and higher background O levels) or interrupting the growth to properly switch the carrier gas and increase the temperature (leading to Mg and eventual contaminations piling up at each interface). Furthermore, In-rich InGaN easily introduces 3D growth after a few nanometers [59].

Whatever approach is chosen, either overdoping, MBE regrowth or InGaN interlayers, all of these introduce a fairly large amount of disorder inside the TJ, leading to a tunneling current mainly supported by the defects. Following this assumption, one may enhance the tunneling current by deliberately introducing deep levels (e.g. doping with carbon, europium, or iron...).

IV. CONCLUSION

We systematically studied the electrical properties, the surface morphology and the Mg, Si, H, C and O concentrations of p-GaN and n-GaN grown by MOVPE at different temperatures for dopant levels

ranging from 10^{18} to 10^{20} cm^{-3} . SIMS results show the C concentration drastically increases from 10^{16} cm^{-3} at 1100°C to about 10^{17} cm^{-3} at 975°C without significantly affecting the electrical properties of both p- and n-type layers. However, O is drastically increased once the Si concentration exceeds 10^{19} cm^{-3} and the growth mode simultaneously changes from 2D to 3D due to Si nanomasking. Overdoping with Mg to 10^{20} cm^{-3} causes inversion domains and defect rich layers which show no longer PL. Thus, at the high Si/Ga and Mg/Ga ratios typically used for p^+ - n^+ tunnel junctions, both type of layers are resistive and present rough surfaces or inversion domains. SIMS analysis of a np-n LED indicates a strong Mg carry over. The pinning of the Fermi level, close to the conduction band in the n-GaN, destabilizes the Mg-H complex ($\text{Mg}/\text{H} \ll 1$) and allows the unintentionally incorporated Mg to efficiently compensate the doping level of the first nanometers of the n-GaN top layer. However, introducing more defective n^{++}/p^{++} layers lead to the desired tunneling. Increasing dopant concentration (and thus defects) reduces the penalty voltage. Based on these results, we conclude that tunnel junctions grown by MOVPE present a poor structural quality and mostly work in defects assisted tunneling.

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