Electronic Supplementary Information

Revealing inhomogeneous Si incorporation in GaN on nanometer scale by electrochemical etching

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Sensitivity of ECE to the presence of defects and to structure orientation

Electrochemical etching is sensitive to the presence of defects, in particular to dislocations. In order to prevent etching from the top surface, it is covered by a compact SiO_2 layer. In case the layer is not perfectly adhering, etching can occur via defects. Figure S1(a) shows the optical microscopy image of a top-surface of a sample after 40 min etching at 4V. Near the grooves, there are brighter areas visible because etching occurs from the grooves towards the middle of the stripes. There is one spot (in the bottom part of the image) where etching occurs via a defect in SiO_2 cap layer.

Electrochemical etching rate depends on structural orientation. However, in our experiment, the etching is defined by sample geometry (top surface is covered by SiO_2) and therefore it proceeds from the grooves, parallel to sample surface along nonpolar directions. Note that when sample orientation is c-plane (0001) GaN, all the *in-plane* directions are nonpolar, including m- and a-directions, i.e. $\langle 1\bar{1}00 \rangle$ and $\langle 11\bar{2}0 \rangle$, respectively. Uniform etching rate along all nonpolar directions is shown on the optical microscopy image presented in Figure S1(b-c). Significant anisotropy in etching rates along +c, -c and nonpolar direction was observed e.g. in S. Mishkat-Ul-Masabih et al., *Applied Physics Letters* 112, 041109 (2018) for a constant doping and bias. Note that authors used another GaN substrate orientation, i.e. $(1\bar{1}00)$, while we use (0001) GaN.

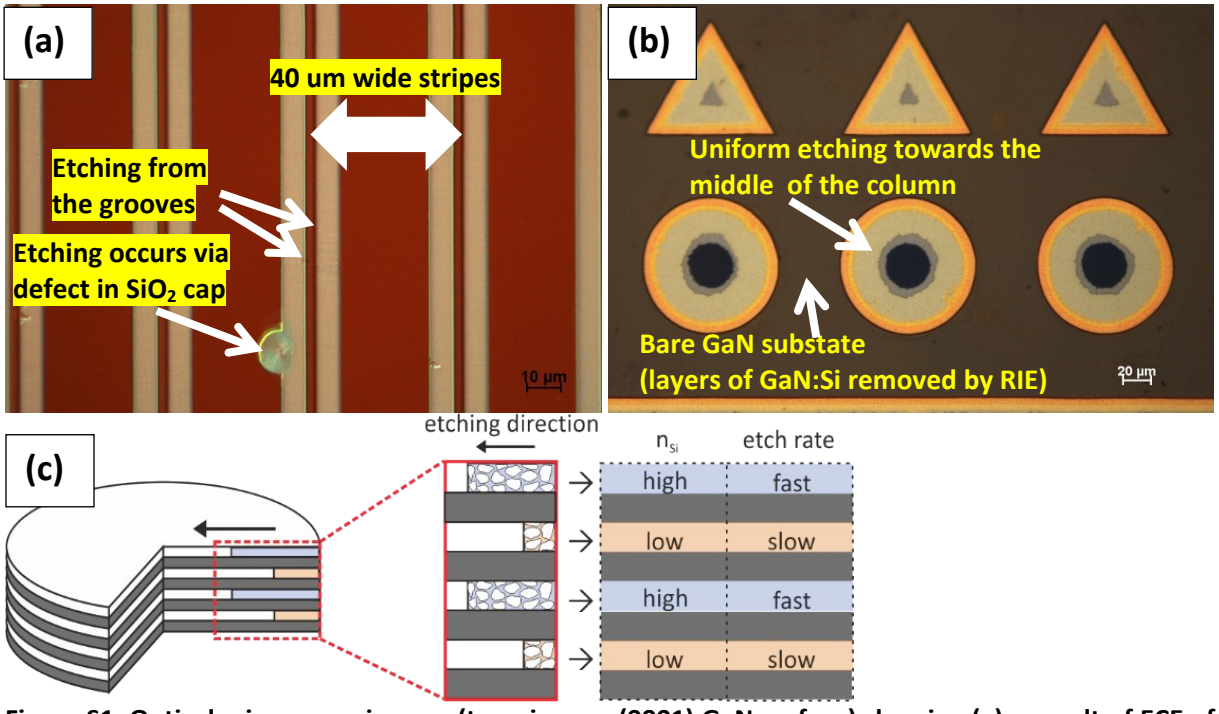

Figure S1. Optical microscopy images (top-view on (0001) GaN surface) showing (a) a result of ECE of 40 min at 4V. Etching from the grooves towards the middle of the stripes is visible. One site where etching via defect was possible is denoted. (b) Triangular and circular columns comprising GaN/GaN:Si layers doped at two different levels after ECE. Bluish layers were etched faster than reddish because they are doped with Si to a higher level. Colors are visible due to formation of porous structure beneath the surface. A schematic picture of a column from (b) is presented in (c). The light passes through different number of porous layers and therefore different colors under the optical microscope are seen.

The presence of defects could also influence etching morphology or perturb incorporation of Si. However, we do not observe fingerprints of that hypothesis. Figure S2(a) and (b) show top-view of GaN:Si layers after ECE for two cases – when layer was grown atomically flat and when it was grown step-bunched. The images show relatively large areas. Please note that we use high-quality bulk GaN substrates of low dislocation density - ~5×10^6 cm^{-2} (this value was confirmed by us recently for the purpose of this study). For such defect density we expect 4 defects on 10x10 μm^2 area AFM/SEM images. Large-area SEM scans of the samples grown with atomically smooth and step-bunched surface confirm no influence of defects on the observed inhomogeneous Si incorporation/etching.

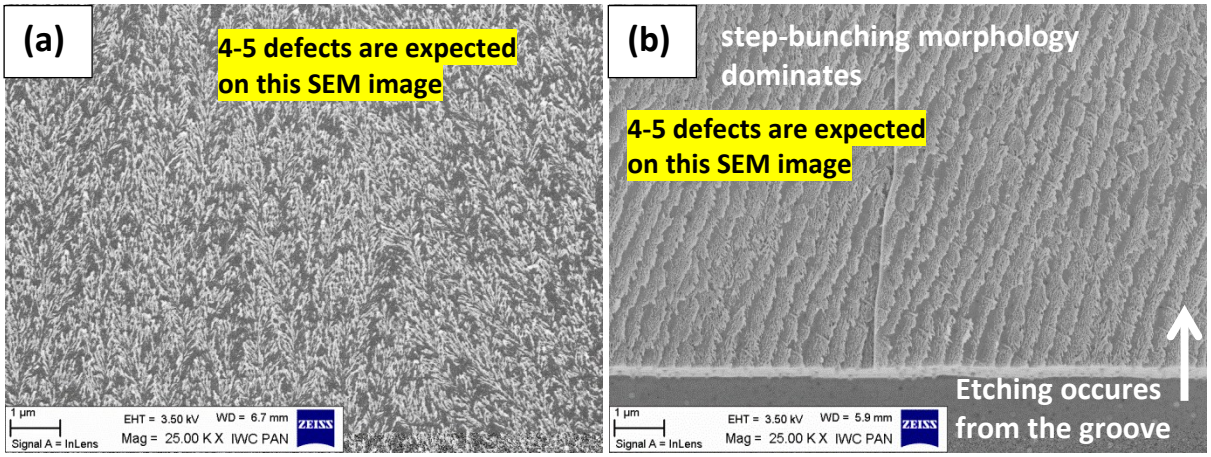

Figure S2. Top-view SEM image of GaN:Si after etching at 4V observed after scratching the sample surface and detaching the top-surface layer for the case when GaN:Si was grown (a) atomically smooth and (b) step-bunched. The area visible on SEM scan is ~12x12µm² (the sample is viewed at 45°). For the given substrate dislocation density, ~4-5 defects are expected on this area.

Surface morphology of undoped GaN

Step-bunching can be induced by the decrease in supersaturation [I. Bryan et al., *Journal of Crystal Growth* 438, 81 (2016)]. In our work it is related to the ratio between metal and nitrogen fluxes Ga/N. However, the presence of Si could influence the surface energy γ, and therefore influence the morphology as well.
In order to verify whether Si doping impacts step-bunching formation, two undoped GaN samples 300 nm thick were grown. The difference was in a growth rate only. Identically as for the GaN:Si layers, the smooth surface was observed for GaN grown at low growth rate and step-bunching for GaN grown at high growth rate. Please, see the results presented in Figure S3, where AFM scans of undoped GaN layers are shown. Therefore we concluded that there is no effect of Si doping on morphology at least for the doping up to 10^{20} cm^{-3}.

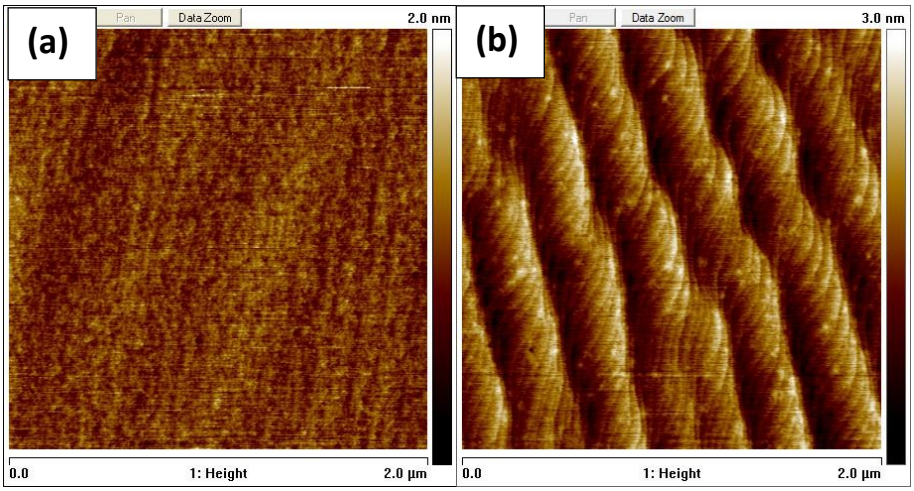

Figure S3. Surface morphologies of 300 nm thick undoped GaN layers grown (a) at low growth rate 0.35 µm/h and (b) high growth rate 0.92 µm/h (identical as the ones used in the article).