

Electronic Supplementary Information

Electronic Supplementary Information for “Core-Shell InGaN/GaN Nanowire Light Emitting Diodes analyzed by Electron Beam Induced Current Microscopy and Cathodoluminescence Mapping”

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Additional experimental details

The InGaN/GaN core-shell organized nanowires have been grown by MOVPE on n-doped GaN/sapphire templates with an SiN mask with sub-micrometer openings. The mask has been patterned by nanoimprint lithography and dry etching. First, an n⁺-doped GaN core is formed (doping concentration of about $1 \times 10^{19} \text{ cm}^{-3}$), then it is laterally overgrown with an n-doped GaN layer with a lower doping concentration ($\sim 5 \times 10^{18} \text{ cm}^{-3}$), one undoped ~ 7 nm thick InGaN quantum well, an AlGaIn electron blocking layer and a p-doped GaN exterior shell (thickness ~ 150 nm, Mg concentration about $\sim 5 \times 10^{19} \text{ cm}^{-3}$). The growth is terminated with a p⁺-doped GaN surface layer to achieve a low contact resistance. This growth procedure results in a homogeneous array of identical vertical nanowires with a density of 5×10^7 nanowire per cm^2 as illustrated in Figure S1 a). The total nanowire height is $2.6 \pm 0.1 \mu\text{m}$ and the diameter is $0.9 \pm 0.05 \mu\text{m}$. The nanowires have a cylindrical shape with a hexagonal cross-section over the lower $2 \mu\text{m}$ with lateral facets

defined by m-planes.¹ By adjusting the growth temperature and fluxes the material deposition on the semi-polar planes is strongly suppressed as seen from a TEM image in Figure S1 b). It is seen that the InGaN quantum well is completely absent on the semipolar planes whereas the thickness of the p-doped GaN shell is strongly reduced down to ~ 20 nm. The electron blocking layer thickness is almost the same on the m-planes and on the semi-polar planes close to 10 nm. The composition of the quantum well and the electron blocking layer has been probed by Energy-dispersive X-ray spectroscopy (EDX). Fig. 1Sd) shows the EDX map of the nanowire lamella cut from the central part of the wire. The In content increases towards the upper part of the quantum well. The In (Al) content in the quantum well (electron blocking layer) measured close to the nanowire base (blue point in Fig 1Sd) and to the top of the m-plane (black dot in Fig 1Sd) is $13.5 \pm 1.5\%$ ($9 \pm 1.5\%$) and $18.5 \pm 1.5\%$ ($19 \pm 1.5\%$), respectively.

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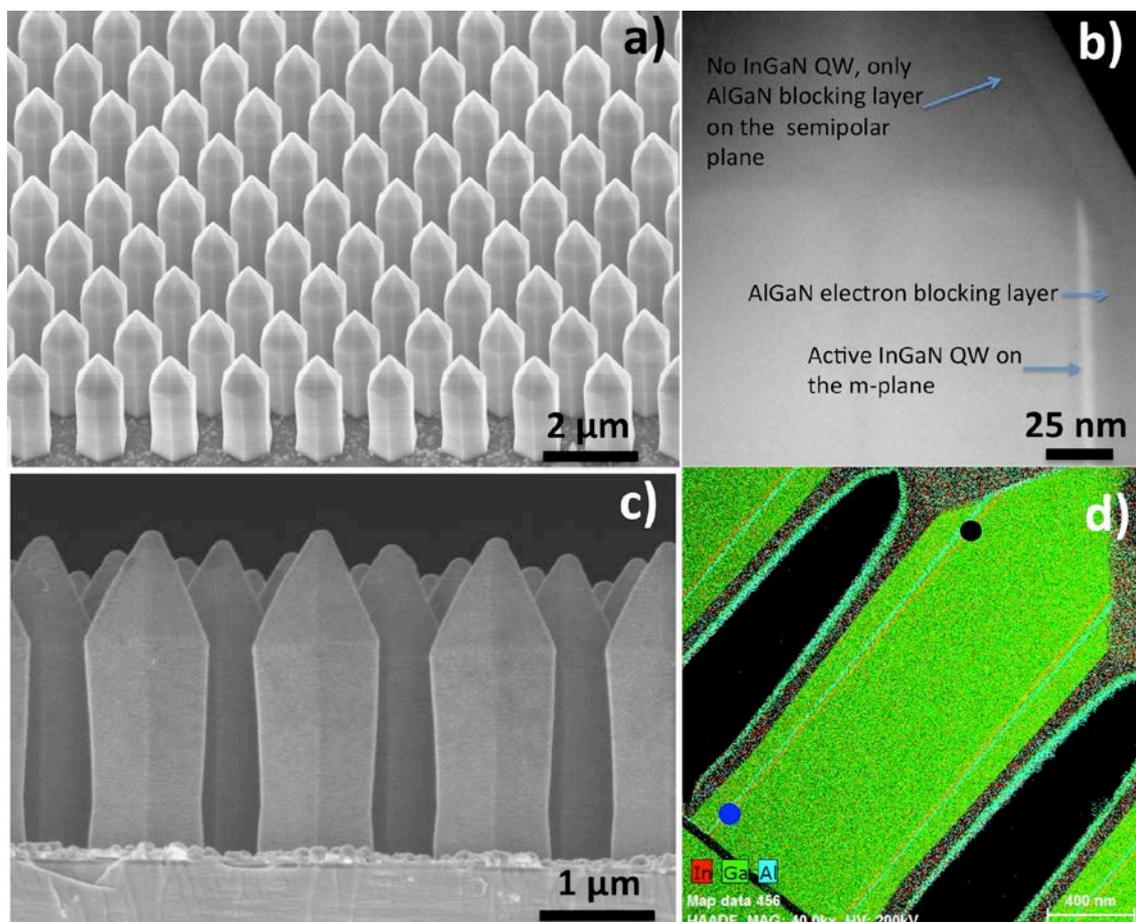


Fig S1. a) Tilted SEM image of as-grown nanowires. b) TEM image showing the active region on the semipolar facet and on the top part of the m plane. c) Cross sectional SEM image of a cleaved nanowire LED. d) EDX mapping of the nanowire cross-section. The blue and black dots show the positions for which the In concentration in the quantum well and the Al concentration in the electron blocking layer is given in the text.

The nanowire array LEDs were fabricated in the form of square mesas containing a large number of parallel-connected nanowires. The mesa size was varied from $300 \times 300 \mu\text{m}^2$ up to $3 \times 3 \text{ mm}^2$. The mesas were defined by optical lithography and inductively coupled plasma dry etching. The top p-type contact was obtained by a conformal deposition of 200 nm of indium tin oxide (ITO) layer onto the nanowires using sputtering deposition and a lift-off. The sample was annealed at 400 °C for 10 min to improve the ITO conductivity. It should be noted that no spin-on glass planarization has been performed, so that the final device has a rough surface. The 2D GaN layer on sapphire was used for bottom n-type contact. Ti/Al/Ti/Au metallization was deposited on the 2D GaN and on the top ITO surface leaving the central part of the mesa open for light extraction. Figure S1 c) displays the cross-section of the fully processed nanowire LED. The evident roughness of the nanowire

sidewalls is due to the ITO layer continuously covering the nanowire array.

Simulation of the top-view EBIC maps

The top-view EBIC signal was simulated using Matlab software in a simple geometrical approximation. The nanowire is represented as a hexagonal cylinder having a hexagonal pyramid on top with the facet inclination angle of 60°. For the cylindrical part, the p-n junction is supposed to be located 150 nm deeper than the m-planes and for the pyramidal part it is placed 20 nm below the semipolar planes, in correspondence with the actual geometry of the nanowires. The ITO thickness was supposed to be 120 nm. The perpendicular extension of the collecting region is supposed to be 150 nm.

The generation volume was approximated with a sphere of a radius defined by the calculated penetration depth partially overlapping with the nanowire as schematized in Figure S2 a). Figure S2 b) illustrates how the spherical approximation has been obtained. The electron trajectories and the penetration depth were calculated using Casino software² and the actual complex excitation volume was replaced by a spherical volume supposing a constant excitation inside the sphere and zero

excitation outside. The presence of the InGaN quantum well and of the AlGaN electron blocking layer has been neglected in Monte Carlo simulations. Calculations for different generation sphere radii and different positions of its center were performed. A geometrical overlap of the generation sphere and of the collecting region was taken as a value of the generated EBIC signal for each point of the calculated top-view map.

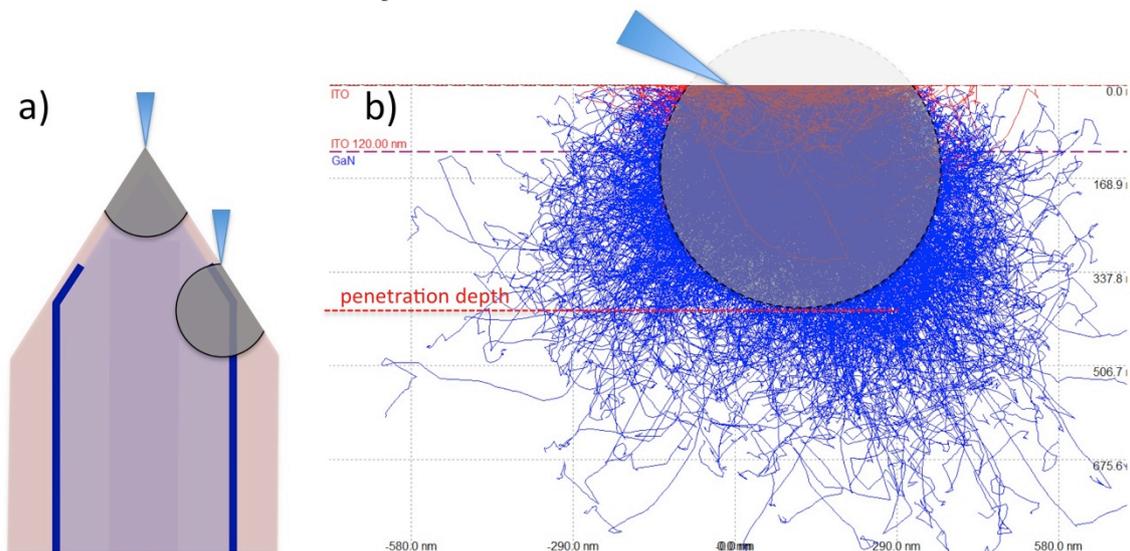


Fig S2. a) Schematics of the nanowire excitation with the electron beam. b) Calculated electron trajectories for 10 kV electron beam impinging the nanowire surface covered with 120 nm ITO layer at an incident angle of 60°. The dashed line shows the penetration depth *i.e.* the distance corresponding to the maximum of the statistical distribution of the depths reached by incident electrons divided by Euler number e .

The p-n junction region was supposed to be homogeneous in the cylindrical part. For the semi-polar planes different extensions of the p-n junction were considered: the p-n junction covering the full semi-polar plane surface, the p-n junction reduced to the lower trapezoidal part of the semi-polar facets (as schematized in Fig S2 a)) and no p-n junction on the semi-polar facet. The best agreement with the experimental results was found for a p-n junction covering one third of the semi-polar facets.

Cathodoluminescence spectra at different excitation positions

To illustrate the presence of the In gradient in the quantum well, Figure S3 a) displays the CL spectra of a cleaved

nanowire for two different excitation positions shown in panel b): close to the nanowire base and close to the top of the cylindrical part of the nanowire. The CL spectra were collected at room-temperature under $V_{acc}=5$ kV using a Jobin Yvon Spectrometer HR320 coupled with a CCD camera. The spectral range corresponds to the InGaN emission wavelengths. It is seen that for the excitation at the nanowire base, the emission is peaked at 430 nm, whereas for the excitation in the upper part the peak shifts to 480 nm. This confirms that the In content in the quantum well increases toward the nanowire top. An example of a panchromatic CL map of a cleaved nanowire collected at 4K is shown in Figure S3 c) together with its SEM image (panel d)).

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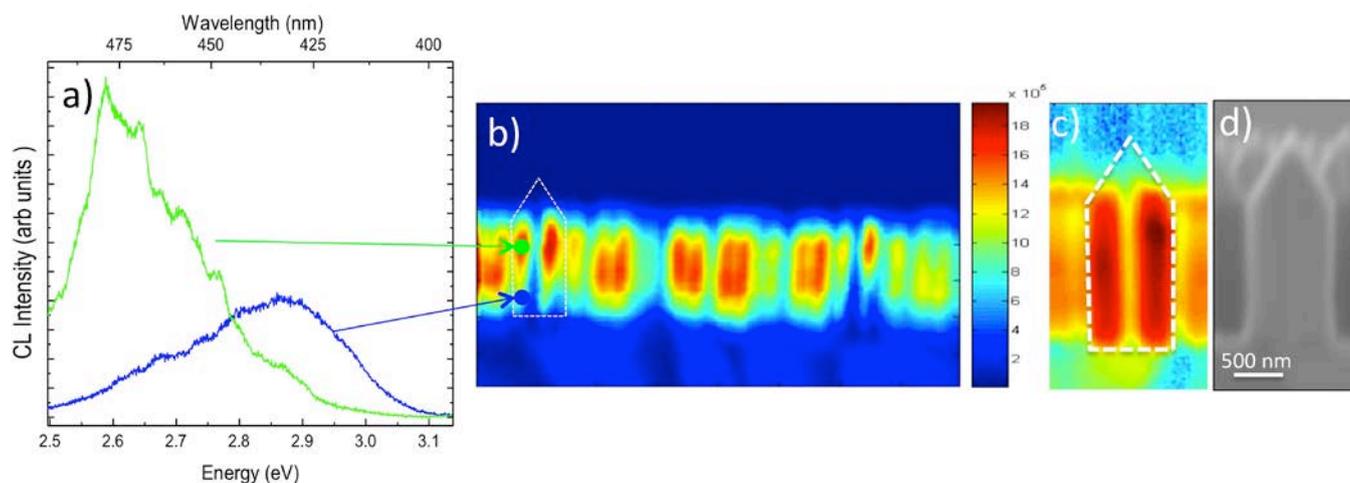


Fig S3. a) Room-temperature CL spectra of a cleaved nanowire for excitation at the base (blue line) and at the top of the cylindrical wire part (green line). b) Room-temperature panchromatic CL map of the sample cross section showing the excitation positions for CL spectra of panel a). c) Low-temperature panchromatic CL map of a cleaved nanowire. d) SEM image corresponding to panel c).

References

- 1 M. Tchernycheva, P. Lavenus, H. Zhang, A. V. Babichev, G. Jacopin, M. Shahmohammadi, F. H. Julien, R. Ciecchonski, G. Vescovi, O. Kryliouk, *Nano Lett.* **2014**, 14, 2456.
- 2 <http://www.gel.usherbrooke.ca/casino/>