Wurtzite GaN nanocolumns grown on Si(001) by molecular beam epitaxy

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Wurtzite single crystal GaN nanocolumns were grown by plasma-assisted molecular beam epitaxy on bare Si(001) substrates. Nanocolumns with diameters in the range of 20–40 nm have no traces of extended defects and they grow aligned along the [0001] direction. Photoluminescence measurements in nanocolumns evidence a very high crystal quality in terms of intense and narrow excitonic emissions. Raman scattering data show that the nanocolumns are strain-free. These results open the way to an efficient integration of optoelectronic devices with the complementary metal oxide semiconductor technology. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2204836]

III-nitrides have a wide potential for a new generation of devices with exceptional performances in electronics applications (high electron mobility transistors, field emission, surface acoustic wave) and optoelectronics (detectors, lasers, light emitting diodes)[1,2] leading the technological edge in many fields, such as lightning, communications, and data storage. One of the most challenging issues is the achievement of a reliable integration of optoelectronic devices with complementary metal oxide semiconductor (CMOS) technology in real integrated circuits. Though sophisticated approaches on heteroepitaxial growth (buffer layer engineering) yielded promising results, no commercial solutions were provided because of complexity, unreliability, high cost, and thermal incompatibility. III-nitrides have been grown on Si(111) at temperatures compatible with Si technology, either by molecular beam epitaxy (MBE) or metal organic vapor phase epitaxy, but still, lattice and thermal mismatches generate a very high density of extended defects in compact (Al)GaN layers. A few attempts to grow GaN compact epilayers on Si(001) aimed either to obtain pure cubic phase GaN (Ref. 3) or wurtzite GaN avoiding cubic inclusions by means of complex buffer structures.[4] None of them were able to achieve high quality, defect-free, single crystal structure layers, neither cubic nor wurtzite.

In the last couple of years a huge effort has been devoted to achieve and to control the growth of III-nitride columnar-shaped nanostructures (nanorods, nanocolumns, nanopillars). Results have shown, so far, an extremely high crystal quality of the (Al)GaN nanocolumns, that are strain-free and have no dislocations or other extended defects, thus, yielding an outstanding emission efficiency. After the growth of self-assembled (Al)GaN and InN nanocolumns on Si(111) by plasma-assisted MBE (PAMBE),[5,6] the achievement of nanocolumnar heterostructures including quantum disks (QDisks) (Ref. 7) and nanocavities with QDisks and Bragg mirrors[8] has been reported. It must be pointed out that nanocolumnar heterostructures can be grown on bare Si(111), without any buffer layer.

It has been shown that III-nitride nanocolumns grown on Si(111) accommodate the lattice mismatch at the heterointerface through a dense network of dislocations that end at the nanocolumn free surface. Once relaxation takes place, the nanocolumn grows keeping its own lattice parameter, a fact that is helped by the nanocolumns high free-surface-to- volume ratio.[9] As a consequence, the nanocolumns should grow without strain and dislocation-free, as it was experimentally observed. In addition, (Al)GaN nanocolumns were grown on a wide variety of substrates [SiC, sapphire, Si(111)].[10,11] Based on these findings, GaN nanocolumns are expected to grow strain-free, defect-free on Si(001) with a single hexagonal crystal structure.

This letter presents the growth of extremely high quality GaN wurtzite nanocolumns on bare Si(001) substrates.

The samples were grown by PAMBE and characterized by scanning electron microscopy (SEM), high resolution transmission electronic microscopy (HRTEM), Raman Spectroscopy (RS), and low temperature photoluminescence (PL).

GaN nanocolumns, 0.6 μm high, were grown on on *axis* ($\pm 0.5-1°$) Si(111) and Si(001) substrates at the optimal temperature used for Si(111) as a starting point in order to compare results from both substrates. Though the optimal temperature to grow on Si(100) may be different, the most important aspect to achieve nanocolumnar growth is the use of strong nitrogen-rich conditions[5] (III/V ratio $\ll 1$). The growth rate was estimated to be around 0.2 μm/h (SEM data in Fig. 1).

The GaN nanocolumn morphology was assessed by SEM. On both, Si(001) (Fig. 1) and Si(111) (not shown) substrates, the nanocolumns are aligned perpendicular to the substrate with a rather uniform distribution of diameters (20–40 nm) and heights.

HRTEM data, obtained with a Jeol JEM 3010 microscope operating at 300 kV, prove that GaN nanocolumns grown on Si(001) are structurally uniform and single crystal-

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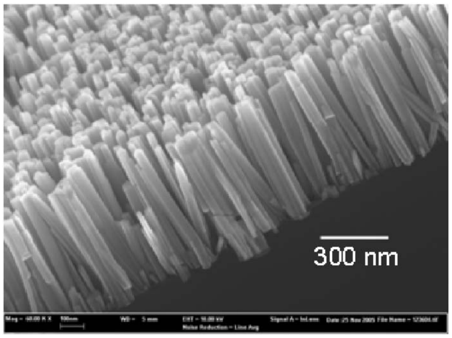

FIG. 1. SEM micrograph of GaN nanocolumns grown by PAMBE on Si(001) substrate.

line, with the growth direction along the hexagonal [0001] axis [Fig 2(a)]. As it was the case of (Ga,Al)N nanocolumns grown on Si(111),[9] no dislocations or other extended defects were detected [the very few stacking faults found are probably related to the occasional coalescence of columns, as shown in Fig 2(b)]. The selected area electron diffraction (SAED) pattern, shown in Fig. 2(c), reflects a texturelike behavior of the nanocolumns, i.e., the GaN(0002) planes are parallel to the Si(002), while the in-plane alignment is not well defined, although few columns show an epitaxial orientation with their [11−20] [1−100] directions parallel to the Si⟨110⟩ axis. The HRTEM micrograph shown in Fig. 2(b) gives an example of a perfect epitaxially aligned hexagonal nanocolumn with GaN(0002)‖Si(002) and GaN[11−20]‖Si⟨110⟩. The interface structure reflects an amorphous layer, most probably SiN, likely formed during the growth due to nitrogen diffusion beneath the nanocolumns through the dislocations network. The study of the degree of columnar twist with respect to the Si substrate surface is still in progress.

Low temperature PL on GaN nanocolumns grown on Si(001) and Si(111) was excited with the 325 nm line of a He–Cd laser, dispersed by a Jobin-Yvon THR 1000 monochromator and detected with a UV-enhanced GaAs photomultiplier and a lock-in amplifier.

PL spectra from GaN nanocolumns grown on Si(001) reveal the same emission lines as those measured on nanocolumns grown on Si(111) (Fig. 3 and Refs. 10 and 12).

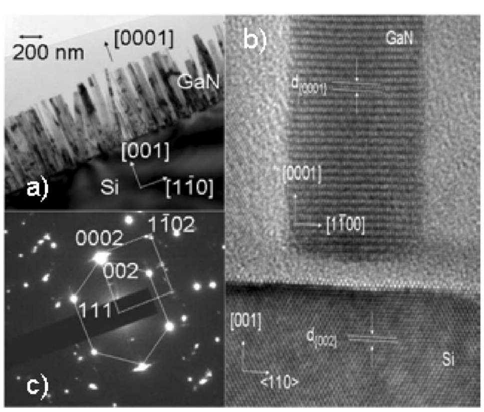

FIG. 2. (a) Cross-sectional TEM image of GaN nanocolumns grown on Si(001). (b) Cross-sectional HRTEM image of a GaN nanocolumn epitaxially aligned to the Si(001) substrate. (c) SAED pattern. The solid and dashed lines indicate the Si[110] and GaN[11−20] zone axis orientations, respectively. All spots in the SAED pattern belong to either Si or hexagonal GaN.

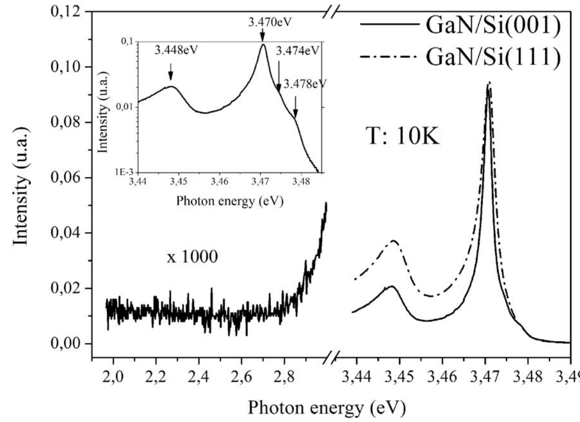

FIG. 3. Low temperature PL spectra of GaN nanocolumns grown on both Si(111) and Si(001) substrates. The inset shows in more detail the dominant near band edge emissions from the GaN nanocolums grown on Si(001) substrate.

Emission lines at 3.470–3.478 eV correspond to excitonic transitions, either donor bound or free. The inset in Fig. 3 resolves up to three excitonic lines at 3.470, 3.474, and 3.478 eV, most probably related to two neutral-donor bound excitons (D^0X) and the free exciton A (FXA), respectively.[11,12] PL emission is dominated by a D^0X line with a full width at half maximum (FWHM) of 2.3 meV, a lower value than that measured on GaN nanocolumns grown on Si(111) (4.3 meV) and far smaller than the corresponding to compact GaN layers grown on Si(111) substrates.[5] Deep level emissions related to structural defects and impurities, such as the *yellow band* centered at 2.3 eV,[13] were not detected (Fig. 3). Similarly, cubic GaN-related emissions, typically observed around 3.3 eV, were not observed.[14] The sharp and intense excitonic emissions and the absence of deep level related emissions point to a very high crystal quality of the GaN nanocolumns grown on Si(001). From the band edge PL emission energies measured, we can conclude that the nanocolumns are strain-free. This is further confirmed by Raman measurements. Data in Fig. 4 show the high energy E_2 Raman mode of GaN nanocolumns grown either on Si(001) or Si(111). In both cases the peak is centered at 566 cm^{-1}, corresponding to fully relaxed thick GaN layers. The width (~5 cm^{-1}) of the Raman lines is the same for both samples and comparable to that of high quality GaN compact samples.[15] This is again indicative of the high crys-

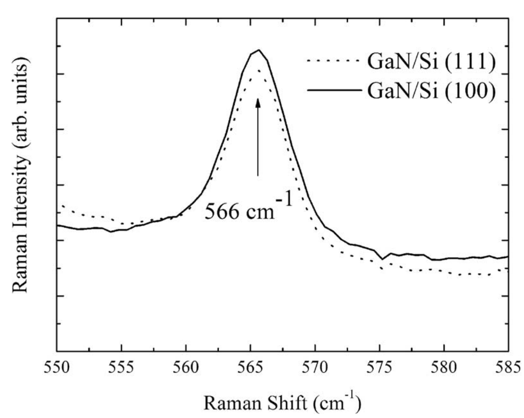

FIG. 4. Room temperature Raman spectra of GaN nanocolumns grown on both Si(001) and Si(111) substrates.

talline quality of the nanocolumns grown on both substrate orientations.

In conclusion, this work demonstrates that pure hexagonal GaN nanocolumns can be grown on Si (001) substrates, with an extremely high crystal quality evidenced by the intense and narrow emission PL lines. The GaN nanocolumns grown on Si(001) substrates are defect-free and strain-free, as it was the case when grown on Si(111) substrates. TEM data demonstrate that the GaN nanocolumns grow along the [0001] direction. These results open the way to an efficient integration of optoelectronic devices with the CMOS technology.

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