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Fabrication of GaN Nano-Towers based Self-Powered UV Photodetector

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Abstract

Fabrication of unique taper ended GaN-Nanotowers structure based highly efficient ultraviolet photodetector is demonstrated. Hexagonal stacked GaN nanocolumnar structure (nanotowers) grown on AlN buffer layer exhibits higher photocurrent generation which significantly enhances its responsiveness towards ultraviolet illumination and leads to outstanding performance of the device. The fabricated detector display low dark current ($\sim 12\text{nA}$), high $I_{\text{Light}} / I_{\text{Dark}}$ ratio ($>10^4$), fast time-correlated transient response ($\sim 433\mu\text{s}$) upon UV (325nm) illumination. A very high photo responsivity of 2.47 A/W in self-powered (zero applied bias) mode of operation is reported. While in photoconductive mode, the responsivity is observed to be 35.4 A/W @ -3V alongwith very high external quantum efficiency ($\sim 10^4\%$), lower noise equivalent power ($\sim 10^{-13}\text{ WHz}^{-1/2}$) and excellent UV-Vis selectivity. Nanotower structure with lower strain and dislocations as well as reduced trap states cumulatively contributed to augmented performance from the device. The utilization of these GaN-Nanotower structures can potentially be useful towards the fabrication of energy-efficient ultraviolet photodetector.

Keywords: Gallium Nitride, Nanotowers, Ultraviolet, Photodetector

Introduction

Gallium nitride (GaN) ultraviolet (UV) photodetectors (PDs) attracted a lot of attention due to their versatility and ability to serve in an extreme environment. Thereby, GaN-material inherits the detectors with high carrier mobility, high saturation velocity, high-temperature sustainability, high breakdown voltages, inertness and hardness towards chemical and radiation respectively [1]. With various advantageous properties, GaN UV-PD can establish its position in numerous applications such as monitoring of combustion, flame detection, ozone detection, space communication, biological sensors: phototherapy, sterilization control, solar irradiance measurement, industrial monitoring, missile plume detection and ultraviolet astronomy etc.[2] So far various UV PD structures such as p- π -n PDs,[3] p-i-n photodiodes,[4] p-n junction photodiodes,[5] Schottky-barrier photodiodes,[6, 7] and metal-semiconductor-metal (MSM) PDs,[8, 9] have been fabricated. The ease of fabrication and rapid time-correlated response ascertained the advantage of Schottky barrier type and MSM structured UV-PDs as compared to the p-n junction. Recent reports also suggest that extremely low dark currents is achieved with MSM based UV PDs.[10] Since, the reduction in dark current is affected by low defect density, thus device performance can directly be correlated with defects generated during the growth of device structure. Simultaneously, these defects induce stress-strain which are accredited to lattice mismatch, and thermal expansion coefficient difference between GaN and the substrate.[11]

Further, in the process of PDs improvisation, nanostructures (NS) have played a significant role due to very low stress-strain induced electrical as well as optical defects that reciprocate its significant impact on detector performance.[12, 13] Moreover, NSs were also recognized as a structure with high surface to volume ratio i.e., higher aspect ratio. The increased surface area in NS provides more absorption sites to incident photons for generating high density of carriers, and lower resistance path which can cumulatively increase the detection capability of the PDs. Therefore, growth of high surface-to-volume ratio and stress relaxed 3D GaN-NSs play a crucial role in the development of highly efficient optoelectronic devices. Recently, it has been reported that GaN-nanocolumnar structure can offers wavelength tunability, low dislocation density and lower lattice related strain and very good response.[14-16] However, the fabricated devices are non-operational in photovoltaic mode i.e. zero applied bias and display very high response time.

In this letter, we report the utilization of AlN buffer layer (800°C) for growth of lower strain and stress relaxed, higher aspect ratio hexagonal stacked GaN- nanocolumnar structure i.e. nanotowers (GaN-NTs) on Si (111) substrate via PAMBE. The impact of buffer on the quality of film is discussed along with the fabrication of UV photodetector. The performance of the fabricated UV-PD is analysed based on the performance parameters such as responsivity, detectivity, noise equivalent power and external quantum efficiency under UV illumination. Besides, the spectral selectivity of the fabricated UV-PD for the wavelength ranging from 200nm to 800nm under variable applied bias has also been assessed.

Experimental

A unique geometry (as shown in figure 1) i.e. GaN-NT arrays have been grown epitaxially via. plasma assisted molecular beam epitaxy (Compact 21 Riber) system. After standard wet chemical etching and thermal cleaning procedure [17] of Si (111) substrate (figure 1(a)), the initial growth of AlN buffer layer has been performed at 800°C for 30 min with Al beam equivalent pressure (BEP) $\sim 6.8 \times 10^{-7}$ torr and growth rate of 3.2 Å/sec (shown in figure 1(b)). Afterwards, a wetting layer of Gallium (Ga) has been deposited for 10 sec at a lower substrate temperature of 666°C that precedes the nucleation process of GaN growth. Through heat energy gained by providing substrate temperature, Ga molecules and N₂ ion active species acquire enough energy at the substrate surface to forms GaN. The growth of GaN-NTs has been performed at 666 °C substrate temperature for 210 min with Ga BEP of 2.8×10^{-6} torr. To fulfil the essential requirement for nanocolumnar GaN growth, nitrogen-rich condition (III/V ratio <1) was maintained by adjusting N₂ flow up to 3 sccm with 400 W RF-plasma power with growth rate 3.2 Å/sec. Thereafter, the growth conditions of closely packed GaN-NCs (shown in figure 1(c)) has been altered to the tapered ended nanotower-like structure by gradually reducing the Ga-flux from 2.8×10^{-6} to 1.5×10^{-6} torr for next 600 sec (shown in figure 1(d-e))

Figure 1 (a-e) A schematic growth diagram of grown GaN NTs on Si (111) substrate with sandwiched AlN buffer layer in between.

The real-time monitoring of GaN-NTs/Si (111) growth has been performed by reflection high electron diffraction (RHEED) using in-situ STAIB electron gun (12 keV). The morphological studies of grown GaN-NTs have been carried out by using Field-emission scanning electron microscopy (FEI Verios 460L FEGSEM) and Atomic force microscopy (AFM) (Multimode-V Veeco). The structural quality of the grown structure was examined by High-Resolution X-ray Diffraction (HRXRD) (PANalytical X'Pert PRO MRD System) using $\text{CuK}\alpha 1$ radiation ($\lambda = 1.5406 \text{ \AA}$). For analysing the optical properties, room temperature (RT) photoluminescence (PL) measurement was carried out by using FLS980 D2D2 (Edinburg) system with 325nm excitation source (Hd-Cd Kimmon laser) and Raman spectroscopy was carried out in backscattering configuration, using a 514 nm argon-ion laser. The grown GaN-NTs were utilized to fabricate UV-PD in MSM structure by depositing metal electrodes of Au ($\sim 200\text{nm}$)/Cr ($\sim 10\text{nm}$) using thermal evaporation system. Photoresponse measurements were carried out using a probe station setup (S10 Triax probe station with Keithley 2401 acquisition unit) on the fabricated device with $1.18 \times 10^{-3} \text{ cm}^2$ active area. The spectrometer is equipped with a focused laser source ($\lambda = 325 \text{ nm}$ with the utilized power density of 2.5 mW/cm^2 to 184 mW/cm^2).

Results and Discussion

The growth of GaN-Nanotower structure is carried out on reconstructed Si (111) 7x7 and in-situ monitored by RHEED technique (figure S1, supplementary information). The crystallinity of grown structure was evaluated by HRXRD 2θ - ω scan and its peak positions (figure S2; supplementary information) were utilized to calculate the strain via lattice constant estimation using Bragg's law.[18] The calculated value of strain in the grown structure is realized to be 0.25% which confirms strain relaxation in the grown nanotowers. Further, to analyse the dislocation density, HRXRD omega scans along (0002) and (10-12) plane of diffraction have been performed as shown in figure 2(a). The FWHM values of omega scan along (0002) and (10-12) were found to be 0.39° and 0.84° attributing to screw dislocation density $\sim 1.91 \times 10^9 \text{ cm}^{-2}$ and edge dislocation density $\sim 2.33 \times 10^{10} \text{ cm}^{-2}$, respectively. These dislocation densities are found to be lower in comparison to the GaN NTs structure grown on Si (111) substrate with thicker AlN buffer layer (665 nm) at lower growth temperature (762°C).[16]

Figure 2 (a) HRXRD omega scan along (0002) & (10-12) plane of diffraction, (b) Planar view FESEM image (inset: shows a single NT), (c) Cross-sectional FESEM image of GaN NTs.

The surface morphology of the GaN-nanotowers has been probed by FESEM (as shown in figure 2(b)) where the planer view of high density closely packed GaN-NTs with hexagonal shapes nanocolumns was shown with tapered ended geometry. Inset of figure 2(b) demonstrates a high magnification image of layer by layer growth of single GaN-NT. Figure 2(c) displayed the cross -sectional view of the novel hexagonal GaN-NTs having length of $\sim 4.168 \mu\text{m}$ grown along the direction perpendicular to the substrate with AlN-buffer layer ($\sim 580\text{nm}$) sandwiched between them. The notable layered structure is having stacking of uniform hexagonal GaN layers up to the length of $\sim 4\mu\text{m}$. Afterward, lowering of Ga flux resulted in a gradual decrease in the surface areas of stacking of GaN layers which help in formation of unique nanotowers like structure ended with a tip of width: $\sim 100\text{nm}$. The distribution density of grown NTs is calculated to be $3.5 \times 10^{10} \text{ cm}^{-2}$. It was also observed that the high quality AlN buffer layer grown at higher substrate temperature (800°C) resulted in better structural and morphological properties in comparison to the structure grown at AlN buffer grown at lower substrate temperature[19, 20] Besides this, the increment in surface area is evaluated in concurrence with FESEM & AFM measurements which confirm the enhancement from $9 \mu\text{m}^2$ to $15.6 \mu\text{m}^2$ from planar to taper ended GaN-NTs morphology.

Figure 3 (a) RT-PL spectra and (b) Raman spectra of as-grown GaN-NTs.

The grown GaN-NTs with enhanced surface area consequently allow more photon absorption sites to generate a large number of photo-excited electron-hole pairs (EHPs). These EHPs emit high-intensity photoluminescence upon recombination which was reflected by intense near band edge emission (NBE) peak at 363.5nm in RT-PL spectra shown in Figure 3(a). The NBE peak is found to be in close agreement with bulk GaN PL emission ($\sim 364\text{nm}$ i.e., 3.40eV). [21] The FWHM value of NBE peak is found to be 8.51nm . The sharp NBE peak

with low FWHM value endorse the good crystalline quality of the grown GaN-NTs.[22] From PL spectra, an additional peak at 540 nm is also observed which is attributed to yellow band emission (YBE) originated due to the generated point defects during the growth.[23, 24] Furthermore, Raman scattering measurement was employed to quantify the stress in the grown structure as shown in figure 3(b). The Raman spectra exhibits, E₂-high mode of grown GaN-NTs located at 567.603 cm⁻¹ which is shifted by ~0.397 cm⁻¹ as compared to the E₂ (high) phonon mode of bulk GaN (568 cm⁻¹).[21] The observed red-shift in the E₂ (high) position specifies of tensile stress in the grown GaN-NTs.[25, 26] The value of tensile stress[18] is estimated to be 0.092 GPa which is significantly lower as compared to 3D GaN-NSs and 2D GaN films[27-29] which can be attributed to lower defect states. Further, it has been proposed that these defects can have an adverse impact on electrical as well as the optical property of the GaN-based UV PD.[30] So, the lower defects attributes to lower trap states, hence the grown GaN-NTs having lower defect density can significant be useful for fabrication of efficient optoelectronic devices.

Figure 4(a) displays RT current-voltage (I-V) characteristic of fabricated GaN-NTs based UV-PD under dark as well as 325nm UV-laser illumination (3mW optical power) with applied bias voltage sweep of $\pm 3V$. The schematic image of the fabricated GaN-NT array based device is shown in the figure 4(b). The difference in dark and light I-V characteristics of the fabricated device leads to photodetection aspect. As revealed by I-V-plots, the self-channelization of light current [inset figure 4(a)] without applied bias voltage (zero bias) viz. photovoltaic mode designates the fabricated detector as self-powered UV-PD. The presence of non-identical end tips of the grown GaN-NT structure lead to form non-homogeneous Au/GaN interfaces between Au-metal electrodes and GaN-NTs which exhibits two Schottky natured electrodes with different barrier heights. The variation in barrier heights creates built-in potential gradient and powered the device to respond even at zero bias voltage.[31]

Figure 4. (a) RT I-V characteristic of fabricated GaN-NTs based UV-PD under dark and 325nm UV illumination with 3mW optical power. Inset: indicate self-powered characteristic at various optical power and bottom right indicate schematic diagram of the fabricated device, (b) Schematic image of the fabricated GaN-NT array based device.

Initially, the transient photo-response behaviour of the fabricated detector has been realized in photovoltaic mode with the variable incident optical power (from 3mW to 13mW) as shown in figure 5(a). The transient photo-response measurement has been performed by measuring inflow current with periodically switching ON (I_{Light}) and OFF (I_{Dark}) the UV laser with variable optical power. The maximum value of $I_{\text{Light}} / I_{\text{Dark}}$ ratio was recorded to be $>10^4$ under 13mW optical power. The analysis demonstrates a very low dark current $\sim 12\text{nA}$, wherein the fabricated UV-PD shows a stable and significant enhancement in light current value from $67.5\mu\text{A}$ to $545\mu\text{A}$ as function of optical power varying from 3mW to 13mW. Further to explore the capability of the detector to detect low power signal, the optical power is reduced from 0.7mW to $180\mu\text{W}$ correspondingly the light current values were observed to be varying from $3.4\mu\text{A}$ to 70.8nA (Figure S3, supplementary information). Conclusively, the fabricated GaN-NTs based UV-PD has shown remarkable enhancement of $\sim 7700\times$ in the photocurrent value from 70.8nA to $545\mu\text{A}$ by varying optical power from $180\mu\text{W}$ to 13mW in the photovoltaic mode (self-power) of operation.

Figure 5 Incident optical power-dependent performance of GaN-NTs UV-PD in photovoltaic mode (a) time-correlated transient photoresponse, (b) Correlation of responsivity and detectivity with varied optical power, (c) Power-law fitted curve in the experimental data of photocurrent, (d) Reversibility (Response time) of the detector under constant 13mW optical power illumination.

In addition the performance of the fabricated device has also been investigated based on their well-known performance parameters like responsivity (R), noise equivalent power (NEP), detectivity (D) and external quantum efficiency (Q.E: η) and reversibility. using the following equations [32] :

$$R = \frac{I_{ph}}{P_{inc} \times A_0} \quad \dots (1)$$

$$NEP = \frac{\sqrt{2eI_d}}{R} \quad \dots (2)$$

$$D = \frac{R \times \sqrt{A_0}}{NEP} \quad \dots (3)$$

$$\eta = \frac{R \times h \times c}{e \times \lambda} \quad \dots (4)$$

Where P_{inc} stands for incident UV photons optical power illuminated on the detector active (A_0), I_{ph} and I_d is the measured photocurrent and dark current, h is the Planck's constant, c is the speed of light, e is elementary charge and λ is incident UV photon wavelength. Figure 5(b) displays that the fabricated GaN-based detector yielded a gradual increment in responsivity from 0.255 A/W to 2.47 A/W and detectivity from 1.8×10^{10} jones to 1.95×10^{11} jones with increasing incident optical power from 180 μ W to 13mW. Remarkably, the determined responsivity and detectivity is found to be significantly very high compared to the existing bare GaN-based UV-PDs operated in photovoltaic or self-power mode of operation. Table 1 consolidates the comparison of performance parameters of other GaN based UV-PD operable in self-powered mode of operation. The analysis also divulges a linear relationship between photocurrent and incident optical power as demonstrated in figure 5(c). The power law has been fitted on the experimental data, $I_{\text{ph}} \propto P^\theta$, where I_{ph} is the photocurrent and θ determines the photocurrent response w.r.t. optical power density. The θ value is determined to be 0.91, which is close to unity reflecting that the carrier trap states between the conduction band edge and the fermi level are very low.[33] Moreover, to test the reversibility of the detectors, the time-correlated transient photoresponse of the fabricated GaN-based UV-PD has been analysed. The rise time (as measured from 10% to 90%) and decay time (from 90% to 10%) under UV on/off condition was measured to be $\sim 433 \mu$ s and $\sim 434 \mu$ s, respectively as shown in figure 5(d). Table 1 also demonstrates a rapid rise/decay time from the fabricated GaN-NTs/AlN/Si heterostructure based UV PD as compared to other reported bare GaN-based PDs. The GaN NTs with lower AlN buffer layer thickness and low dislocation density leads to reduced resistance path for charge transport resulting in efficient photovoltaic operation.[34]

Table 1: Comparison of performance parameters of GaN-based UV-PDs.

Sample	R (mA/W) at 0V	R (mA/W) at applied bias	Rise / Decay time	QE%	Ref.
GaN-NT/Si(111)	2.47×10^3	3.54×10^4 , -3V	433 μ s / 434 μ s	1.35×10^4	This work
GaN-NT/Si(111)	----	8.86×10^4 , -3V	1.25s/ 1.15ms	3×10^4	[19]
GaN-NW/n-Si(111)	----	19, 5V	2.8s/3.7s	6.7	[29]
GaN/Si(111)	----	282, 15V	----	96.71	[27]
GaN-NW/n-Si(100)	----	68, 5V	2.4s/3.1s	24.4	[29]
GaN-MWA/p-Si	131	----	2ms / 2ms	49.98	[35]
GaN/Al ₂ O ₃	104	----		36	[36]
GaN/Al ₂ O ₃	147	----	0.11ms/0.12ms	50.7	[37]
GaN p-i-n/pattern Al ₂ O ₃	190	210, -5V	----	65	[38]
GaN/Al ₂ O ₃	300	----	<0.1s	----	[39]

To analyse the impact of applied bias on the performance of fabricated GaN-based UV-PD, time-correlated transient response of the detector at a fixed optical power of 3mW and variable bias voltage is acquired as shown in figure 6. The investigation revealed an increment in photocurrent values with increasing applied bias i.e., 66.6 μ A (0V), -0.35mA (-1V), -1.025mA (-2V) and -1.73mA (-3V) which is ascribed to enhanced thermionic field emissions. The increment in the photocurrent with respect to applied bias voltage is also reflected in the performance parameters of the fabricated UV-PD. Figure 7(a) displays the increasing trend of responsivity of the device with an increasing value of applied bias voltage from 0V to -3V. The attained responsivity values are 1.79 A/W and 35.4 A/W at 0V and -3V applied bias respectively by exposing the device using 325nm UV source at 3mW optical power. The responsivity value of 35.4 A/W is significantly higher as compared to other bare GaN-based UV-PD as tabulated in Table 1. However, the recently reported GaN-NT based UV-PD [19] demonstrated higher responsivity of 88.6 A/W at -3V, moreover the performance of the devices is limited to photo-conductive mode only with ~300x higher response time. In addition, the present analysis reports optimised AlN buffer with reduced defect density in GaN-NTs. Consequently, the fabricated detector displayed independence of operability in both photovoltaic and photoconductive modes with significantly higher performance with faster transient response. Besides that, another important parameter i.e., the detectivity of the fabricated device is also calculated to be 1.41×10^{11} jones at 0V as and 1.83×10^{10} jones at -3V which divulge that at applied bias, the high dark current reduce detectivity of the device. Furthermore, the quantum efficiency (QE) of the device which is

related to sensitivity and defined as the conversion rate of incident photons to generate charge carriers that contribute to current conduction have also been evaluated. Figure 7(b) demonstrates the impact of variable optical power and applied bias voltage on the quantum efficiency. In photovoltaic mode, the quantum efficiency was observed to be increased from 97.41% at 180 μ W optical power to 946.75% at 13mW optical power. Further, QE values have also been evaluated at a fixed optical power of 3mW and variable bias voltage (0V to -3V) condition where QE increased from 686.39% (at 0V) and reached up to 1.35×10^4 % (at -3V). High value of quantum efficiency (> 100 %) reveals the existence of a photoconductive gain which is greater than unity.[39] Likewise, to measure the sensitivity of the fabricated detector, noise equivalent power (NEP) of the device has been determined.[40] The calculated value of NEP was 2.42×10^{-13} $\text{WHz}^{-1/2}$ (at 3mW, 0V) which elucidates that the fabricated GaN-NTs based UV-PD can detect a signal of power as low as ~ 200 femtowatt with a signal-to-noise ratio of one after the one-half second of averaging.

Figure 6. Bias dependent time-correlated transient photoresponse of GaN-NTs UV-PD at 3mW optical power illumination

Figure 7 (a) Responsivity and detectivity graph w.r.t applied bias voltage, (b) Quantum efficiency (Q.E) of fabricated GaN-NTs UV-PD w.r.t incident optical power and bias voltage.

The spectral selectivity of the fabricated GaN-NTs based UV-PD has also been measured in the range of 200nm to 800nm at variable applied bias from -1V to -3V (figure 8). The obtained data demonstrates that the fabricated detector exhibits noteworthy UV light detection selectivity, which designate the detector to find its application in UV-A spectral range (320-400nm).

Figure 8: Spectral response characteristic of GaN-NTs based UV-PD from 200 to 800 nm under applied bias.

Conclusion

The fabrication of GaN-NT based high-performance and versatile UV-PD with independence of operability (in photovoltaic as well as a photoconductive mode) has been demonstrated where the detector yielded enhanced performance due to unique NTs tapered ended geometry. The closely packed NTs based layered structure formed by stacking of uniform hexagonal GaN layers offers the higher active surface area to the incident UV-photons. Thus, the high performance of fabricated detector using GaN NTs has been ascertained from reduce resistance path for charge transport. Thereby, in self-powered mode of operation, the fabricated UV photodetector showcased significantly higher $I_{\text{Light}} / I_{\text{Dark}}$ ratio ($>10^4$), lower dark current ($\sim 12\text{nA}$), remarkable enhancement in photocurrent (~ 7700 fold) as a function of applied optical power, highest responsivity (2.47 A/W), higher detectivity (1.95×10^{11} jones), quick response time ($\sim 433\mu\text{s}$), very high QE ($\sim 947\%$) and detectability of signal as low as 200 femtowatt. Subsequently, the performance of the fabricated UV-PD has also been analysed at photoconductive mode of operation where significantly higher responsivity $\sim 35.4\text{ A/W}$ at lower bias voltage of -3V is obtained. The fabricated device with efficient operation and fast transit of charge carriers can effectively be utilized for futuristic optoelectronic devices.

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Conflict of Interest: The authors declare that they have no conflict of interest.

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Figures

Figure 1

(a-e) A schematic growth diagram of grown GaN NTs on Si (111) substrate with sandwiched AlN buffer layer in between.

Figure 2

(a) HRXRD omega scan along (0002) & (10-12) plane of diffraction, (b) Planar view FESEM image (inset: shows a single NT), (c) Cross-sectional FESEM image of GaN NTs.

Figure 3

(a) RT-PL spectra and (b) Raman spectra of as-grown GaN-NTs.

Figure 4

(a) RT I-V characteristic of fabricated GaN-NTs based UV-PD under dark and 325nm UV illumination with 3mW optical power. Inset: indicate self-powered characteristic at various optical power and bottom right

indicate schematic diagram of the fabricated device, (b) Schematic image of the fabricated GaN-NT array based device.

Figure 5

Incident optical power-dependent performance of GaN-NTs UV-PD in photovoltaic mode (a) time-correlated transient photoresponse, (b) Correlation of responsivity and detectivity with varied optical power, (c) Power-law fitted curve in the experimental data of photocurrent, (d) Reversibility (Response time) of the detector under constant 13mW optical power illumination.

Figure 6

Bias dependent time-correlated transient photoresponse of GaN-NTs UV-PD at 3mW optical power illumination

Figure 7

(a) Responsivity and detectivity graph w.r.t applied bias voltage, (b) Quantum efficiency (Q.E) of fabricated GaN-NTs UV-PD w.r.t incident optical power and bias voltage.

Figure 8

Spectral response characteristic of GaN-NTs based UV-PD from 200 to 800 nm under applied bias.

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