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ORIGINAL RESEARCH ARTICLE

Enhanced Photoresponse of a Self-Powered Gallium Nitride Photodetector via Sequentially-Deposited Gold Nanoparticles for Sustainable Optoelectronics

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Abstract
It is becoming crucial to design/fabricate eco-friendly, sustainable electronic and photonic devices to minimize the carbon footprint for future systems. In this study, we have demonstrated a steady photoresponse enhancement of the self-powered GaN ultraviolet photodetector (GaN-UVPD) via sequentially deposited gold nanoparticles (Au NPs) under 254, 302, and 365 nm UV light exposure. The AuNP-deposited GaN-UVPD exhibited excellent responsivity of 0.65 A/W and detectivity of 6.51×10^{12} cm.Hz$^{1/2}$ W^{-1} under 302 nm UV light without any external power. Moreover, the sensitivity of the device increased from 1.98×10^{6}% to 3.32×10^{6}% following Au nanoparticle deposition. Additionally, the plausible mechanisms for the self-powered and Au nanoparticle-induced photoresponse enhancement have been discussed. In brief, the high-performance photoresponsivity of our self-powered GaN-UVPD could find many useful applications in sustainable energy and eco-friendly optoelectronic devices.

Keywords Self-powered photodetectors · GaN-UVPD · Au nanoparticles · plasmonics · sustainable energy

Introduction

Ultraviolet photodetectors (UVPD) have attracted tremendous attention in environmental, industrial, military, and biological applications such as optical communications, pharmaceutical and chemical analysis, environmental sensing, flame detection, biomedical electronics, missile detection, and space communications.[1,2] Photodetectors capable of operating without any external power source can solve the energy requirement along with simplifying and reducing the cost of overall device fabrication. Conventional photodetectors need external power sources to operate, which is not desirable for smart sensor systems and devices. However, self-powered photodetectors can harvest energy from the environment enabling small, portable, cost-effective, and multifunctional devices for future optoelectronic applications.[3–5] Gallium nitride (GaN, $E_g = 3.39$ eV) with high carrier mobility, high saturation velocity, good-temperature sustainability, high breakdown voltage, chemical inertness, and radiation hardness has positioned itself for many optoelectronic device applications.[6]

GaN ultraviolet photodetectors (GaN-UVPDs) have shown many promising advances in terms of their low dark current, high sensitivity, responsivity, and detectivity. For example, one recent study reported that the responsivity of a GaN based metal-semiconductor-metal (MSM) photodetector following the application of gold (Au) nanoparticles (NP) increased from 3.89 to 45.99 mA/W at zero bias.[7] Similarly, the detectivity jumped from 3.43×10^{9} cm.Hz$^{1/2}$ W^{-1} Jones to 30.5×10^{9} Jones following the Au nanoparticle application at zero bias. In another study, a GaN photodetector with Au and silver (Ag) nanoparticles yielded a detectivity of 2.4×10^{12} Jones along with rise and fall times of 160 ms and 630 ms, respectively, at 0.1 V.[8] A different study reported an increase in sensitivity after the incorporation of platinum (Pt) onto a GaN nanowire photodetector, with sensitivity increased from 971% on the bare GaN nanowire to 9975% on the Pt-GaN nanowire under 380 nm UV light at 5 V bias.[9] The same study also reported a decrease in rise time from 17.5 to 1.1 s and decay time from 6.2 to 0.65 s with the

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application of Pt nanoparticles.[9] Although there has been some research done on self-powered photodetectors, it is still very limited compared to conventional photodetectors. Moreover, self-powered photodetectors can be building blocks for individual wireless devices, which are very desirable in some applications including electronic skins (e-skin) and Internet of Things (IoT). Nevertheless, there exists much room for improvement in performance and fabrication cost of the self-powered GaN-based photodetectors.

This paper presents a systematic investigation of the sequentially deposited gold nanoparticles (AuNPs) at various deposition times (no NP, 5-s Au deposition, 10-s Au deposition, 15-s Au deposition, 20-s Au deposition) on the performance of a self-powered (zero power consumption) GaN UV metal–semiconductor–metal photodetector under 254, 302, and 365 nm UV light exposures. It is important to note here that Au nanoparticles have a large number of easily polarizable conduction electrons, which is a general prerequisite for preferential interaction with electromagnetic fields and the generation of nonlinear optical phenomena.[10,11] Moreover, Au nanoparticles attract attention due to their excellent chemical inertness and ease of synthesis. Meanwhile, MSM photodetectors are preferred devices for optoelectronics due to their low capacitance unit area (enables high-speed operation), simplicity of fabrication, and compatibility with field-effect transistor technology. The detectivity increased from 3.89×10^{12} Jones to 6.51×10^{12} Jones following the 20-s Au deposition on the GaN surface under 302 nm UV light at zero bias. Moreover, the responsivity went from 0.39 A/W to 0.65 A/W under the same circumstances (67% increase). Consequently, this work will contribute for developing high-performance self-powered UV photodetectors that can potentially replace traditional high-energy-consuming UV detection and communication systems.

Experimental Details

The GaN-UVPD was fabricated on a Mg-doped GaN ($1.0\times10^{20}/cm^3$) grown on a sapphire (0001) substrate. The wafer (6×6 mm) was cleaned via a standard solvent cleaning method with 5-min acetone sonication followed by 5-min methanol sonication in a beaker. To wrap up the cleaning process, the wafer was rinsed in deionized water (DI) for 2 min and finally dried with nitrogen. A physical mask was used to pattern a gap with dimensions of 1.8 mm×60 µm. The asymmetrical Au electrodes were deposited via a sputter coater system (Agar Scientific) for 60 s. During the Au deposition process, the sputter chamber was pumped down and purged with Argon four times to improve the purity of the Au electrodes. To investigate the influence of the gold nanoparticles on the performance of the photodetector, the surface of the produced GaN-UVPD was coated with gold for 5 s, 10 s, 15 s, and 20 s with a deposition rate of 90 nm/min. Following each Au deposition step, the GaN-UVPD was heated at 300 °C for 5 min to transform the deposited gold film into nanoparticles before making optoelectrical measurements. The surface morphology and the crystalline structure were investigated using scanning electron microscopy (SEM, PhenomWorld XL) and x-ray diffraction (XRD). A sourcemeter unit (Keithley 2634B), which was connected to a probe station and controlled by a LabVIEW program, was used to carry out optoelectrical measurements. These measurements were done in a dark room via a UV lamp (UVLMS 38) with wavelengths of 254 nm, 302 nm and 365 nm and power densities of 1.8 mW/cm^2, 1.6 mW/cm^2, and 1.5 mW/cm^2, respectively.

Results and Discussion

Figure 1 shows an XRD pattern of GaN grown on a sapphire substrate. The diffraction peaks in the spectrum were indexed to a hexagonal wurtzite crystal structure with GaN (0002) at 34.58° and GaN (0004) at 72.85°. Also, the diffraction peak at 41.68° is from the sapphire (0006) plane. After structural characterization of the GaN was complete, the device fabrication was carried out. Figure 2a displays a schematic layout of the self-powered GaN-UVPD with measurement setup. The asymmetrical Au electrodes are separated by a 60 µm gap. Figure 2b shows an SEM image of the GaN-UVPD surface with Au electrodes.

Figure 2c shows an SEM image of the plain GaN surface before any fabrication process. In order to investigate the influence of Au nanoparticles on optoelectrical properties of the GaN-UVPD, the surface of the device was coated with Au for 5 s, 10 s, 15 s, and 20 s, and subsequently heat treated at 300 °C for 5 min. After each Au deposition and heat treatment cycle, the measurements were repeated under the same conditions. Figure 2d demonstrates an SEM image

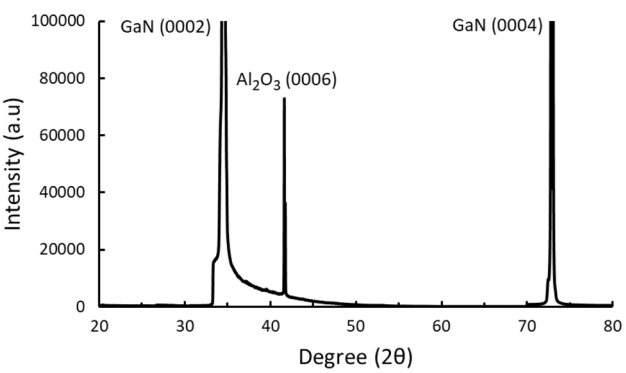

Fig. 1 XRD pattern of the single crystalline GaN grown on a sapphire substrate.

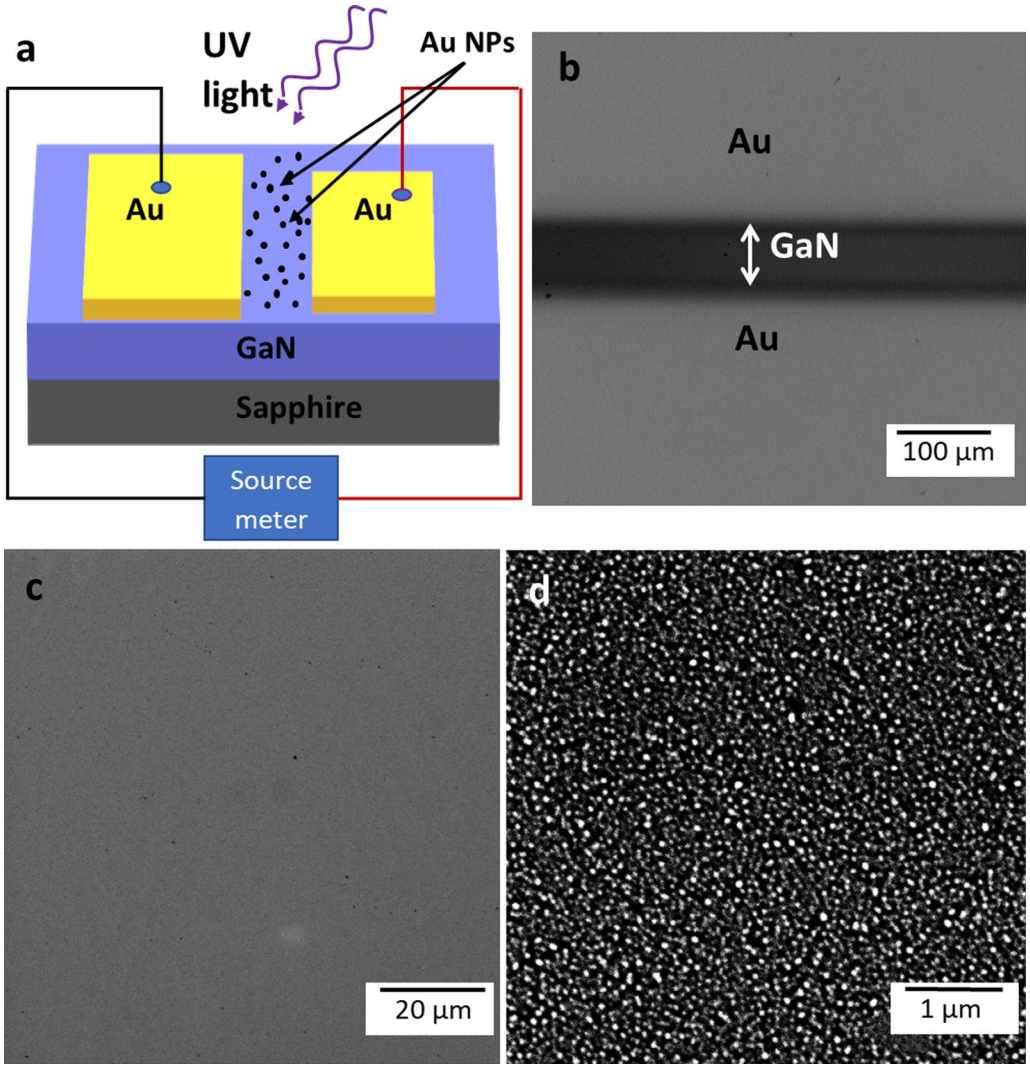

Fig. 2 (a) Schematic illustration of the GaN-UVPD with Au nanoparticles; (b) SEM image of the GaN-UVPD with 60-µm gap between the Au electrodes; (c) SEM image of the plain GaN surface before any fabrication process; (d) SEM image of the GaN-UVPD-Au20s exhibiting uniform distribution of Au nanoparticles on the surface of the GaN-UVPD.

of the GaN-UVPD surface with Au nanoparticles, which are formed by a very thin layer of Au deposition (20 s) and subsequent heat treatment at 300 °C for 5 min. The deposited Au film is cracked and forms Au nanoparticles during the heat treatment process due to the difference in thermal expansion coefficients between Au and GaN. The average size of the Au nanoparticles is about 37 nm. The surface density of the Au nanoparticles (Au-NPs in the active area / total active area of the GaN-UVPD) is calculated as 11.76%. The device was designated as GaN-UVPD-Au20s with 20-s Au deposition, and this labeling will be used in the following sections of this manuscript.

In order to examine the optoelectronic properties of the device, the GaN-UVPD was tested under various UV light exposures of 254 nm, 302 nm, and 365 nm. Figure 3a shows the current vs. time (*I-t*) graphs of the plain GaN-UVPD under various UV lights. Measurements were taken at zero bias with 10 s on–off cycles. The photocurrent is about 130 nA, 400 nA, and 650 nA, with UV light exposure of 365 nm, 254 nm, and 302 nm, respectively. The dark current of the device is very low (3.4×10^{-11} A). The photo-to-dark current ratio of the device under the 302 nm UV light exposure is about five times that of the 365 nm UV light for the plain GaN-UVPD. Following the plain GaN-UVPD measurements, the GaN-UVPD was tested with sequentially deposited Au nanoparticles. Figure 3b shows the current vs. time graphs of the 20-s

Au deposited GaN-UVPD. In fact, the photocurrent values increased substantially from 130 to 150 nA for the 365 nm UV light (15% increase), from 400 nA to 550 nA for the 254 nm UV light (37.5% increase), and from 650 nA to 1120nA for the 302 nm UV light (72% increase) due to the

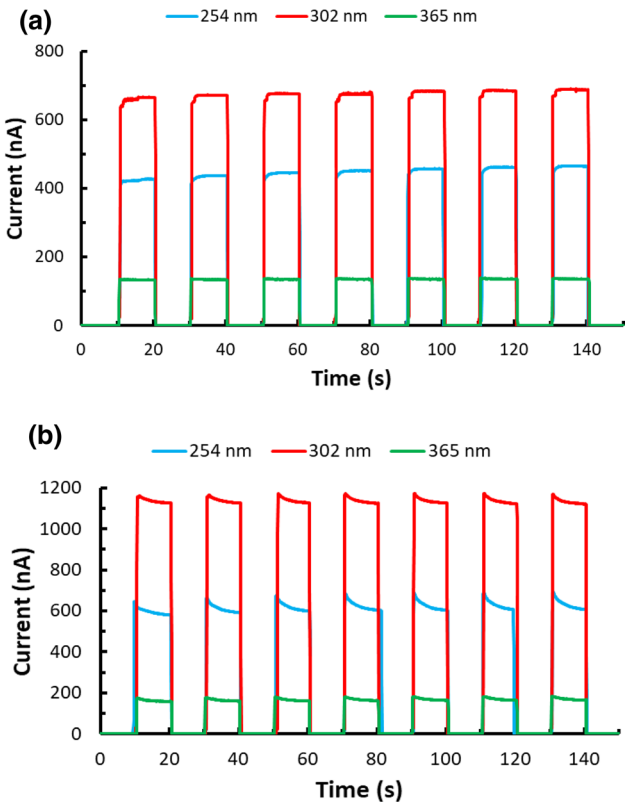

Fig. 3 (a) Current–time characteristics of the plain GaN-UVPD at zero bias under various UV light exposures with 10-s cycles; (b) current–time graphs of the 20-s Au deposited GaN-UVPD under various UV light exposure (254 nm, 302 nm, 365 nm) at zero bias.

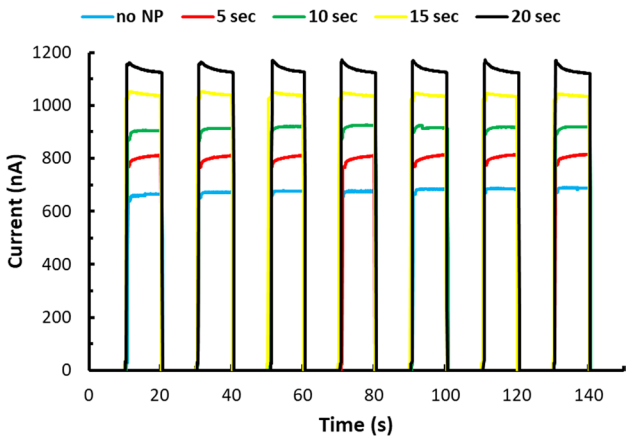

Fig. 4 The current–time responses of the GaN-UVPD at different Au deposition times (plain surface, 5 s, 10 s, 15 s, 20 s) under the irradiation of 302 nm UV light at zero bias, showing a steady increase in photocurrent with Au deposition.

application of Au nanoparticles for the GaN-UVPD-Au20s. While considerable enhancements have been achieved through each light source, the greatest impact was observed with the 302 nm UV light. It was reported in a recent study that the responsivity of a GaN-based photodetector to ultraviolet light with wavelength around 300 nm is higher than that of 254 nm.[12] Thus, the investigation of the influence of Au nanoparticles at various deposition times on the GaN-UVPD focused on the 302 nm UV light. It is important here to discuss two plausible mechanisms of the photocurrent improvement due the application of Au nanoparticles to the GaN-UVPD surface. First, the Au nanoparticles on the surface of GaN can enhance the scattering of the incident photons and thus lead to absorption of the photons by the semiconductor. The Au nanoparticles can couple a larger fraction of the incident light into the GaN surface due to the enhanced near-field coupling.[13,14] Second, the increase in photocurrent due to Au nanoparticles could be due to localized surface plasmon resonance (LSPR), where the nanoparticles increase light absorption of semiconductors due to the strong local field enhancement around the Au nanoparticles. When nanoparticles are much smaller than the wavelength of incident light, they can act as a "nanoantenna" and store the incident light energy in localized surface plasmon mode.[15] These localized surface plasmons lead to a large number of photoexcited carriers that are transferred into the GaN, resulting in a significant increase in photocurrent of the photodetector upon exposure to the UV light. Thus, Au nanoparticles on the GaN surface enhance UV light absorption leading to an increase in the number of carriers.

Figure 4 presents the current–time responses of the GaN-UVPD at different Au deposition times (plain surface, 5 s, 10 s, 15 s, 20 s) under the irradiation of 302 nm UV light at zero bias. The photocurrent values are about 650, 810, 880, 1040, and 1120 nA for the Au deposition times of 0 s, 5 s, 10 s, 15 s, and 20 s, respectively. In an attempt to rank the photocurrent changes with Au deposition times relative to the plain GaN-UVPD, 5-s Au deposition yields 25% increase, 10-s Au deposition 35% increase, 15-s Au deposition 60% increase, and finally 20-s Au deposition yields 72% increase. Thus, the photo-to-dark current ratio increases from 1.98×10^4 to 3.32×10^4 as the Au deposition time increases. Even though we have observed a steady increase in photocurrent until 20-s Au deposition, some studies reported that excessive nanoparticle coverage of the device surface can hinder the light absorption, leading to a reduction in the performance of the photodetector.[16] It was reported in a very recent study that the performance of a gold nanoparticle-decorated ZnO photodetector degraded as the size of the gold nanoparticles increased.[17] This will be further investigated in our future study.

Sensitivity (S) is an important parameter used in characterizing the optical properties of a photodetector and is expressed by, $S=(I_{on}–I_{off})/I_{off})\times 100$ (1), where I_{on} is the photocurrent and I_{off} is the dark current. Since the dark current of our device is very low (3.4×10^{-11} A), the difference between the sensitivity and the I_{on}/I_{off} ratio is

very minimal. The corresponding sensitivity values are 1.98×10^6%, 2.38×10^6%, 2.70×10^6%, 3.05×10^6%, and 3.32×10^6% for Au deposition times of 0 s, 5 s, 10 s, 15 s, and 20 s, respectively. While assessing the performance of photodetectors, it is particularly important to study two parameters, namely responsivity and specific detectivity. Both are good indicators in understanding the characteristics of a photodetector. Responsivity (R) is defined as the photocurrent generated per unit power of the incident light on the effective area,[18,19] and is expressed by $R = I_{ph}/(P_\lambda\ x\ A)$ (2), where I_{ph} is the net photocurrent (I_{on}–I_{off}), P_λ is the light intensity, and A is the effective exposed area. In fact, the responsivity increased from 0.39 A/W (plain GaN-UVPD) to 0.65 A/W (20-s Au deposited GaN-UVPD) under 302 nm UV light irradiation at zero bias, respectively.

Specific detectivity (D^*) is described as the ability of a photodetector to detect small optical signals.[18] As the detectivity increases, the capability of the device to sense smaller optical signals increases as well. The specific detectivity is expressed by, $D^* = (R\ x\ A^{1/2})/(2\times q\ x\ I_{off})$ (3), where R is the responsivity, A is the effective exposed area, q is the electronic charge, and I_{off} is the dark current. Similarly, the detectivity also improved with the increase of Au deposition times ranging from 3.89×10^{12} Jones to 6.51×10^{12} Jones for the plain GaN-UVPD and 20-s Au deposited GaN-UVPD under 302 nm UV light irradiation at zero bias, respectively. Table I summarizes the important results in terms of I_{on}/I_{off} ratio, sensitivity, responsivity, and specific detectivity of the GaN-UVPD at various Au deposition times under 302 nm UV light exposure at zero bias.

Figure 5 displays the responsivity and detectivity values of the self-powered photodetector as a function of Au deposition time at 0 V under 302 nm UV light revealing significant increases with the application of Au nanoparticles to the device surface. In fact, the responsivity increased from 0.39 to 0.65 A/W for the Au deposition times of 0 s and 20 s, respectively. Moreover, the detectivity also improved from 3.89×10^{12} Jones to 6.51×10^{12} Jones for the plain GaN-UVPD and 20-s Au deposited GaN-UVPD at zero bias, respectively. These results show the efficiency and the ability of sensing small optical signals of our self-powered UV photodetector decorated with uniformly distributed Au nanoparticles.

Here, it is essential to discuss the self-powering mechanism of the MSM GaN-UVPD device. The traditional MSM photodetectors with two symmetric back-to-back Schottky contacts on a planar surface require an external power source to generate photocurrent. In fact, MSM devices fabricated with two different electrode materials (one ohmic and one Schottky contact) can operate without any external power. Nevertheless, these MSM devices suffer from poor performance and complex fabrication steps. These drawbacks can be overcome by using an asymmetric pair of planar electrodes enabling efficient separation of the photogenerated carriers due to a difference in electric field built in each Schottky junction.[20,21] Figure 6 illustrates the schematics of the self-powering mechanism based on the asymmetric contact area under UV light. When the contact areas of GaN and Au are different from each other at each junction, it will lead to different depletion widths between Au and GaN junctions, thus creating a difference in the built-in electric field between the two junctions. Hence, an asymmetrical metal-semiconductor-metal configuration will appear. The larger junction area electrode will have a large number of

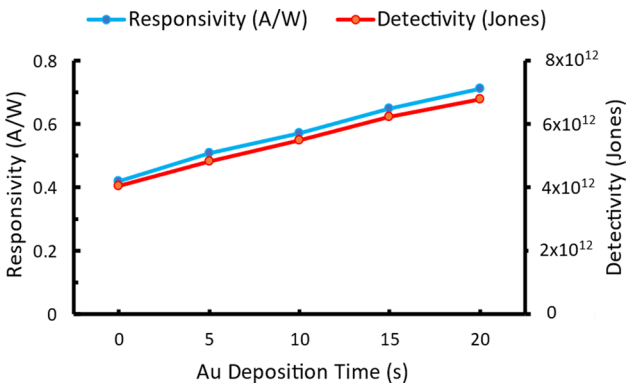

Fig. 5 The responsivity and detectivity plots of the self-powered GaN-UVPD as a function of Au nanoparticle deposition time, i.e., plain GaN-UVPD, GaN-UVPD-Au5s, GaN-UVPD-Au10s, GaN-UVPD-Au15s, and GaN-UVPD-Au20s.

Table I Comparison of the I_{on}/I_{off} ratio, sensitivity, responsivity, and detectivity of the GaN-UVPD at various Au deposition times (0 s, 5 s, 10 s, 15 s, 20 s) under 302 nm UV light at 0 V

302 nm UV light at 0 V				
Au deposition time, s	I_{on}/I_{off} Ratio	Sensitivity, %	Responsivity, A/W	Detectivity, $cm.Hz^{1/2}W^{-1}$
No. Au NPs	1.98×10^4	1.98×10^6	0.39	3.89×10^{12}
5	2.38×10^4	2.38×10^6	0.47	4.66×10^{12}
10	2.70×10^4	2.70×10^6	0.53	5.30×10^{12}
15	3.05×10^4	3.05×10^6	0.60	5.98×10^{12}
20	3.32×10^4	3.32×10^6	0.65	6.51×10^{12}

holes lowering the Schottky barrier more as compared to the smaller Au-GaN junction. As a result, an uneven built-in potential will occur across the photodetector.

When the photodetector is connected to an external circuitry and exposed to the UV light, the photogenerated carriers are driven towards the electrodes leading a photocurrent due to the difference in Schottky barrier heights between the two junctions without any applied bias voltage.[20]

Finally, Table II positions the performance of our GaN-UVPD compared to some other similar recent studies.[22–26] For example, one study reported a GaN photodetector with aresponsivity of 0.20 A/W and a detectivity of 3.10×10^{9} Jones under 325 nm UV illumination and at 0 V bias.[23] Another self-powered study of the Ga_2O_3 UVPD with Sn alloyed nanostructures reported responsivity and detectivity values of 2.9×10^{-5} A/W and 1.79×10^{9} Jones, respectively.[24] Additionally, our very recent study on GaN-UVPD with randomly distributed Au nanoparticles (drop-cast from a solution) yielded responsivity and detectivity values of 1.32×10^{-2} A/W and 1.07×10^{11} Jones, respectively at 1 V.[26]

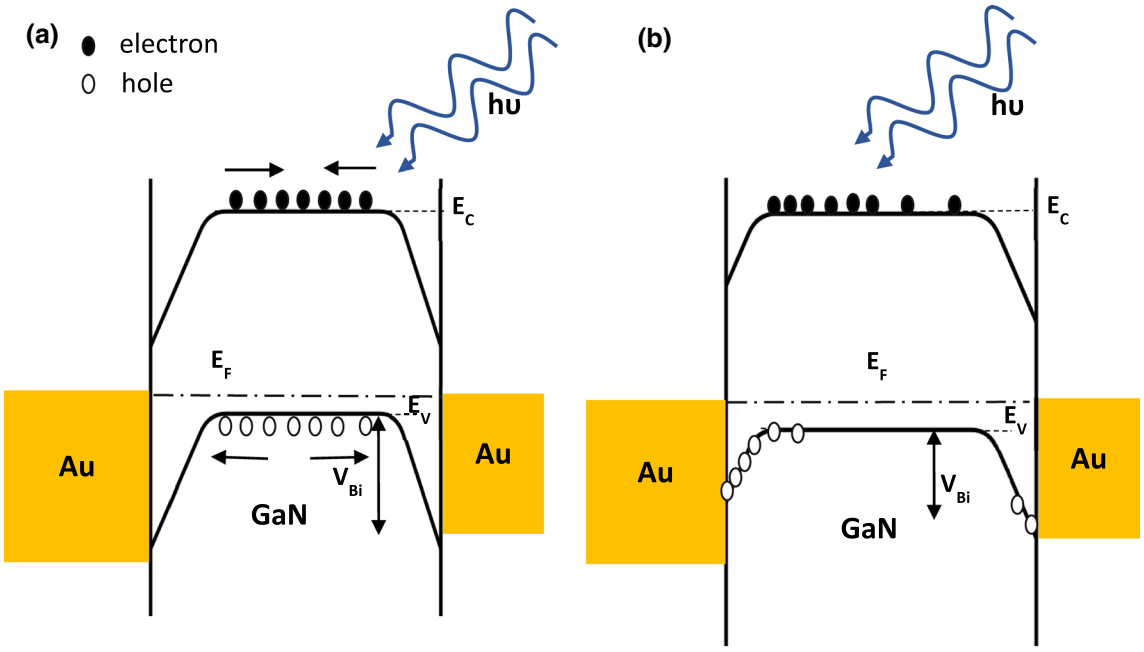

Fig. 6 Schematic band diagrams of the self-powered MSM GaN-UVPD at 0 V under UV light: (a) initial illumination, (b) elapsed time (E_C: conduction band; E_V: valence band; E_F: Fermi level of the system; $V_{B\text{-}in}$: built-in potential voltage).

Table II Comparison of the ultraviolet photodetector performance parameters including responsivity and detectivity of several externally powered and self-powered photodetectors

Material System	Wavelength, nm	Bias voltage, V	Responsivity, A/W	Detectivity (Jones)	Reference
Non-polar GaN UVPD	325	5	0.34	1.24×10^{9}	22
GaN UVPD on Si substrate	325	0	0.20	3.10×10^{9}	23
GaN UVPD on Si substrate	325	1	1.50	0.91×10^{9}	23
Ga_2O_3 UVPD-Sn Alloyed Nanostructures	302	0	2.9×10^{-5}	1.79×10^{9}	24
Ga_2O_3 UVPD-Sn Alloyed Nanostructures	302	2	3.98×10^{-3}	2.40×10^{11}	24
ZnO UVPD-0.8 mg CdMoO4-Au NP	350	5	0.19	–	25
GaN-UVPD with 15 µL of Au NPs	365	1	1.32×10^{-2}	1.07×10^{11}	26
GaN-UVPD No NPs	302	0	0.39	3.89×10^{12}	This work
GaN-UVPD-Au5s	302	0	0.47	4.66×10^{12}	This work
GaN-UVPD-Au10s	302	0	0.53	5.30×10^{12}	This work
GaN-UVPD-Au15s	302	0	0.60	5.98×10^{12}	This work
GaN-UVPD-Au20s	302	0	0.65	6.51×10^{12}	This work

In fact, we have dramatically improved the performance of our GaN-UVPD via employing a more uniform distribution of Au nanoparticles on the GaN surface with responsivity and detectivity values of 0.65 A/W and 6.51×10^{12} Jones at 0 V bias, respectively. In consequence, our improved and easy approach would enable the fabrication of high-performance self-powered UV photodetectors that can potentially replace traditional high-energy-consuming UV detection systems.

Conclusions

We have successfully presented a systematic investigation of the sequentially-deposited gold nanoparticles (AuNPs) at various deposition times (0 s, 5 s, 10 s, 15 s, 20 s) on the performance of a self-powered GaN-UVPD under 254, 302, and 365 nm UV light exposures. The GaN-UVPD exhibited substantial enhancements in detectivity, responsivity, and sensitivity with Au nanoparticle application. For instance, the detectivity increased from 3.89×10^{12} Jones to 6.51×10^{12} Jones following the 20 s Au deposition on the GaN surface under 302 nm UV light at zero bias. Moreover, the responsivity improved from 0.39 A/W to 0.65 A/W under the same circumstances (67% increase). Furthermore, the sensitivity rose from 1.98×10^{6}% to 3.32×10^{6}. Also, the device showed very good speed with the rise and recovery times of 8 and 48 ms, respectively. The improvement in detectivity, responsivity, and sensitivity from the Au nanoparticles is likely due to a strong local field enhancement around the Au nanoparticles and enhanced scattering of the incident photons leading to a strong light absorption around the Au nanoparticles resulting an increase in number of photoexcited carriers. In short, the high performance photoresponsivity of our self-powered GaN-UVPD could find many useful applications in sustainable energy and eco-friendly optoelectronic devices.

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

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