Self-Powered and Spectrally Distinctive Nanoporous Ga$_2$O$_3$/GaN Epitaxial Heterojunction UV Photodetectors

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Herein, self-powered spectrally distinctive UV photodetectors (PDs) based on nanoporous epitaxial Ga$_2$O$_3$/GaN heterojunctions grown by metalorganic chemical vapor deposition (MOCVD) are reported. The nanoporous structures are formed by a novel self-reactive etching (SRE) method, which significantly enhances the device performance. PDs with different porosities are fabricated and compared. In the self-powered mode, the PD with the highest nanopore porosity exhibits the best performance, with an ultralow dark current of 0.04 nA, a fast photoresponse speed, and a high responsivity of 43.9 mA W$^{-1}$ (35.8 mA W$^{-1}$) under 254 nm (365 nm) illumination, which is 800% higher than that of the reference device without the porous structure. Furthermore, opposite current polarities are observed in the PDs under different illumination spectra, the mechanism of which is explained by the carrier transportation using the device band diagrams. This phenomenon can be used to distinguish different incident spectra, opening the door to more new applications. This work represents one of the first spectrally distinctive self-powered UV PDs based on nanoporous Ga$_2$O$_3$/GaN heterojunctions and significantly benefits the development of multifunctional UV PDs.

1. Introduction

UV photodetectors (PDs) have been widely used in civil and military applications, such as optical communication,[1–3] high-voltage corona detection,[4,5] high-flux flame sensing,[6] and missile plume detection.[7] Nowadays, high-performance UV PDs have to meet the “5S” requirements, i.e., high sensitivity, high speed, high signal-to-noise ratio, high spectrum selectivity, and high stability.[8,9] To meet the “5S” requirements and improve the detection system integration capability, heterojunction structures have been proposed to realize high-performance UV PDs, such as GaN/Ga$_2$O$_3,[12,13]$ AlGaN/GaN,[14,15] GaN/MgZnO,[16] and GaN/ZnO.[17,18] Furthermore, with the increasingly popular applications in Internet of Things (IoT), wireless technology, secure communication, and space detection,[19] another important emerging requirement for UV PDs is ultralow power or even no external power. Self-powered UV PDs have garnered significant research interest in these applications[20,21] due to their low energy consumption. Several groups recently demonstrated self-powered heterojunction UV PDs.[22–24] Chen et al.[25] reported a self-powered PD based on NiO and a ferroelectric (FE) material (Pb$_{0.95}$La$_{0.05}$Ti$_{0.46}$O$_3$) with a responsivity of 0.18 mA W$^{-1}$ and a low dark current of 1.3 pA. The device performance enhancement was mainly due to the combined FE depolarization field and the built-in field.

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RESEARCH ARTICLE

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In this work, we demonstrate self-powered spectrally distinctive nanoporous Ga$_2$O$_3$/GaN heterojunction UV PDs. The devices take advantage of the unique optical properties of Ga$_2$O$_3$ (e.g., a suitable bandgap of 4.5–4.9 eV\(^{28}\) and a large absorption coefficient of \(10^5\) cm\(^{-1}\) near the band edge\(^{29}\)) and GaN (e.g., high electron drift velocity). The nanoporous structures were fabricated using a novel self-reactive etching (SRE) method, and the effects of these structures on the PD performance were investigated. The results show that the nanoporous Ga$_2$O$_3$/GaN heterojunction can provide a new method for self-powered spectrally distinctive UV PDs with high performance.

2. Results and Discussion

Figure 1a shows the device structure of the fabricated nanoporous Ga$_2$O$_3$/GaN heterojunction UV PDs after the SRE and metallization. The interdigital electrodes were used to increase the output photocurrent and optimize the frequency characteristics of the devices.\(^{30}\) As shown in Figure 1b, the effective detection area \((S)\) of the PD was \(4.92 \times 10^{-4}\) cm\(^2\), which is significantly increased by the nanoporous structures and leads to stronger light absorption. The Ga$_2$O$_3$/GaN heterojunction is expected to offer the self-powering capability and dual-band detection, which will be characterized in the following sections.

As shown in Figure 2a, the X-ray diffraction (XRD) \(2\theta\) scan of the Ga$_2$O$_3$ film indicates that the film mainly consisted of \(\varepsilon\)-phase Ga$_2$O$_3$. It was reported that the \(\varepsilon\)-phase was beneficial for improving the uniformity of the nanopores.\(^{31}\) The high-resolution XRD rocking curve of the Ga$_2$O$_3$ film in Figure 2b shows that the Ga$_2$O$_3$ film had decent crystal quality with a full width at half maximum (FWHM) of 0.62°. To reduce the effect of surface roughness on the formation of Ga droplets for SRE, the growth conditions of Ga$_2$O$_3$ were optimized. Figure 2c shows an atomic force microscopy (AFM) image of the Ga$_2$O$_3$/GaN heterojunction. The film had a low surface roughness of 1.86 nm in a \(5 \times 5\) \(\mu\)m\(^2\) scanning area. These results indicate that high-quality Ga$_2$O$_3$/GaN epitaxial layers were obtained with a smooth surface morphology and good material quality grown by metalorganic chemical vapor deposition (MOCVD) on sapphire substrates.

Figure 3 shows the material characterizations of the nanoporous Ga$_2$O$_3$/GaN films after the SRE process. During the MOCVD growth, the triethylgallium (TEGa) flow rate was varied from 0, 10, 50, to 90 sccm, and the resulting films are denoted as "G0," "G1," "G2," and "G3," respectively. With increasing TEGa flow rate, the porosity and density of nanopores decreased, whereas the diameters of the nanopores increased. This is because TEGa flow rates can influence the amounts of available Ga atoms on the film surface, resulting in different densities of Ga droplets during the SRE. It has been reported that the available Ga atoms on the film surface are closely related to the size of the Ga droplets but have no impact on their density. With more available Ga atoms on the film surface, the average diameter of the Ga droplets increased but their density remained almost

![Figure 1. a) Schematic structure of the nanoporous Ga$_2$O$_3$/GaN heterojunction PDs. b) Optical microscopy image of the fabricated PDs.](image)

![Figure 2. a) Powder XRD of the Ga$_2$O$_3$/GaN heterojunction. The powder XRD of GaN/Al$_2$O$_3$ is also shown for comparison. b) High-resolution XRD (001) rocking curve of the Ga$_2$O$_3$ film. c) AFM image of the Ga$_2$O$_3$ film surface.](image)
unchanged. Therefore, the Ga droplets formed under high TEGa flow rates tended to be larger in size, but the spacing between neighboring Ga droplets was smaller. This led to Ostwald ripening (i.e., increasing size of large droplets at the expense of small droplets) and cluster migration during the annealing stage, leading to the decrease in the density and porosity of nanopores. Figure 3e shows that the nanopore width and depth were \( 80 \) and \( 100 \) nm, respectively. Figure 3f shows the reflectivity spectra of the four nanoporous films G0, G1, G2, and G3. The lowest reflectivity was observed in G1 with the highest porosity in the deep UV region. This suggests that G1 has the highest UV absorption, which is suitable for UV PDs. In addition, Fabry–Perot resonance was also observed in all samples when the wavelength was larger than 254 nm.

The nanoporous Ga\(_2\)O\(_3\)/GaN heterojunction PDs based on G0, G1, G2, and G3 are denoted as “PD0,” “PD1,” “PD2,” and “PD3,” respectively, where PD0 serves as a reference. The current–voltage (I–V) characteristics of the nanoporous PDs and the reference device PD0 were investigated under illumination with wavelengths of 254 nm (corresponding to the \( \beta\)-Ga\(_2\)O\(_3\) band edge) and 365 nm (corresponding to the GaN band edge) and in the dark. When the four PDs were exposed to 254 and 365 nm illumination, the photocurrent \( I_{\text{photo}} \) of these PDs showed different degrees of enhancement compared with the reference PD0, as shown in Figure 4a,b.

Under the 254 nm illumination, PD1 had the largest \( I_{\text{photo}} \) of 24.71 \( \mu \)A at reverse bias, which is more than 13 times that of PD0 (\( I_{\text{photo}} = 2.11 \mu \)A) at \(-5\) V. Under the 365 nm illumination, the largest \( I_{\text{photo}} \) (\( I_{\text{photo}} = 138.4 \mu \)A) of PD1 was more than 69 times that of PD0 (\( I_{\text{photo}} = 1.98 \mu \)A). These results are consistent with the highest porosity and density of nanopores in the G1 film. This is because PD1 fabricated on the G1 film has the largest effective area for light absorption, resulting in the highest \( I_{\text{photo}} \).

The typical rectifying characteristics of nanoporous Ga\(_2\)O\(_3\)/GaN heterojunction PDs were observed in the dark. The dark current \( I_{\text{dark}} \) of PD0, PD1, PD2, and PD3 were 2.69, 0.04, 0.10, and 0.07 nA at 0 V, respectively. The photo–dark current ratios (PDCRs) were calculated at 0 V by the equation

\[
\text{PDCR} = \frac{I_{\text{photo}}}{I_{\text{dark}}} \cdot 100\%
\]

Figure 3. SEM images of nanoporous Ga\(_2\)O\(_3\) thin films under different TEGa flow rates: a) 0 sccm, b) 10 sccm, c) 50 sccm, and d) 90 sccm. e) Cross-sectional SEM image of G1. f) Reflectivity spectra of nanoporous Ga\(_2\)O\(_3\) films etched under different TEGa flow rates.

Figure 4. I–V curves of the nanoporous Ga\(_2\)O\(_3\)/GaN heterojunction PDs: a) 254 nm illumination with an intensity of 2400 \( \mu \)W cm\(^{-2}\) and in the dark; b) 365 nm illumination with an intensity of 30 mW cm\(^{-2}\) and in the dark.
PCDR = $I_{\text{photo}}/I_{\text{dark}}$. Under the 254 nm illumination, the PCDRs of PD1 and the reference PD0 were 272.15 and 4.60, respectively, and those values were 6408.44 and 26.42, respectively, under the 365 nm illumination. The short-circuit currents of PD1 were 12.38 and $-291.52\, \text{nA}$ under 254 and 365 nm illumination, respectively. These results indicate that these PDs are self-powered, and the nanoporous structure can dramatically enhance the self-powering capability of the Ga$_2$O$_3$/GaN heterojunction PDs.

Responsivity ($R$) of a PD represents its sensitivity to incident light and is calculated by $R = (I_{\text{photo}} - I_{\text{dark}})/(P \cdot S)$, where $P$ is the light intensity and $S$ is the effective area. As shown in Figure 5a,b, a significant enhancement in responsivity was obtained in PD1 compared with the reference PD0, and PD2 and PD3. Specific detectivity ($D^*$) describes the smallest detectable signal, which is calculated by $D^* = R/(2eJ)^{1/2}$, where $e$ is the electron charge and $J$ is the dark current density. Figure 5c,d shows the $R$ and $D^*$ curves of PD1 as a function of power intensity under different illuminations. The largest values of $R$ and $D^*$ were 43.9 mA W$^{-1}$ and $2.7 \times 10^{11}$ Jones under the 254 nm illumination with an intensity of $170\, \mu\text{W cm}^{-2}$, while those values were $35.8\, \text{mA W}^{-1}$ and $2.17 \times 10^{11}$ Jones under the 365 nm illumination with an intensity of $47\, \mu\text{W cm}^{-2}$. The linear dynamic range (LDR) can be determined by the equation $\text{LDR} = 20 \log(P_{\text{max}}/P_{\text{min}})$, where $P_{\text{max}}$ is the largest input light power and $P_{\text{min}}$ is the minimum detectable light power.$^{[34]}$ The LDR of PD1 was 23 dB under the 254 nm illumination and 56 dB under the 365 nm illumination. It should be noted that due to the testing setup limitations, the LDR of the devices in this work are slightly lower compared with previous reports. The external quantum efficiency (EQE), defined as the number of electrons detected per photon and given by $\text{EQE} = \text{Rhc}/\lambda e$, is another critical parameter to evaluate the performance of PDs. The EQE of PD1 was 5.1% (6.7%) under the 254 nm (365 nm) illumination at 0 V.

Figure 6 shows the transient response of PD1 and the reference PD0 under the 254 and 365 nm illumination at different applied biases. With increasing reverse bias, both $I_{\text{dark}}$ and $I_{\text{photo}}$ of the devices became larger. The increase in $I_{\text{dark}}$ of the devices is attributed to the release of more carriers from the oxygen vacancy traps and the enhanced drift velocity.$^{[35]}$ The increase in $I_{\text{photo}}$ of the devices is due to a larger built-in electric field in the heterojunction and the resulting faster separation of the electron–hole pairs under high reverse biases. The rise time $\tau_r$ of PDs is defined as the time for the current rising from 10% to 90% of the peak value, and the decay time $\tau_d$ of PDs is defined as the time for the current dropping from 90% to 10% of the peak value. In Figure 6c–f, $\tau_r$ and $\tau_d$ were extracted for PD1 and the reference PD0. Under the 365 nm illumination, the response time ($\tau_r/\tau_d$) of PD1 was 0.01/0.05 and 0.02/0.18 s at 0 and $-2\, \text{V}$, respectively, and those of PD0 were 0.02/0.05 and 0.03/0.02 s, respectively. When in the self-powered mode, the response time ($\tau_r/\tau_d$) of PD1 is comparable to or slightly better than that of the reference PD0. Under the 254 nm illumination, the response time ($\tau_r/\tau_d$) of PD1 was larger than that of the reference PD0. This is due to the electrons and holes released from the interface states that were introduced during the SRE process. Furthermore, in the self-powered mode, an interesting $I_{\text{photo}}$ polarity reversal phenomenon was observed when the PDs were under different illuminations. Under the 254 nm illumination (Figure 6a,d), the $I_{\text{photo}}$ of PD1 and that of the reference PD0 were positive, whereas the $I_{\text{photo}}$ of the devices was negative under the 365 nm illumination (Figure 6b,e). The physical mechanism for this current polarity reversal phenomenon will be explained in detail later. Table 1 summarizes the device
performance of the nanoporous Ga$_2$O$_3$/GaN heterojunction UV PDs and previously reported dual-band UV PDs.

Figure 7 shows the band diagrams of the Ga$_2$O$_3$/GaN heterojunction PDs to explain the photoelectric conversion process and the opposite polarity of $I_{\text{photo}}$ under different illumination wavelengths. Figure 7a shows the energy band diagrams of GaN and Ga$_2$O$_3$ before contact. When the Ga$_2$O$_3$/GaN heterojunction is formed, the electrons from Ga$_2$O$_3$ flows toward GaN until the Fermi levels in both materials line up, forming an equilibrium band diagram (Figure 7b) with built-in potential barriers for carriers. The majority carriers of Ga$_2$O$_3$ and GaN are electrons (due to donor-type oxygen vacancies) and holes, respectively. When the Ga$_2$O$_3$/GaN heterojunction is forward biased, the built-in potential barriers are reduced. The majority carriers together with photogenerated carriers overcome the barrier and flow to the opposite sides (Figure 7c), forming electron

**Table 1.** Comparison of the device parameters of the reported dual-band PDs.

| PDs                        | $\lambda$ [nm] | $R$ [%A W$^{-1}$] | Bias [V] | $I_{\text{dark}}$ [pA] | $D$ [Jones] | Self-powered | Ref. |
|----------------------------|----------------|-------------------|----------|-------------------------|-------------|--------------|------|
| β-Ga$_2$O$_3$/Cu core–shell microwire | 254            | 8.46              | 0        | 8.5                      | 7.75 x 10$^{11}$ | Yes          | [34] |
|                            | 365            | 1.36              |          |                         | 1.25 x 10$^{11}$ |              |      |
| p-GaN/Ga$_2$O$_3$          | 254            | 28.44             | 0        | 1.2                      | 6.17 x 10$^{10}$ | Yes          | [35] |
|                            | 365            | 54.43             |          |                         | 1.23 x 10$^{11}$ |              |      |
| β-Ga$_2$O$_3$/black phosphorus (BP) | 250            | 88.5              | –3       | 2.4                      | –            | No           | [37] |
|                            | 1024           | 1.24              |          |                         |              |              |      |
| Ga$_2$O$_3$/p-CuScN        | 254            | 13.3              | 5        | 1.03                     | 9.43 x 10$^{11}$ | Yes          | [38] |
|                            | 365            | 1.5 x 10$^{-3}$   |          |                         | –            |              |      |
| Ga$_2$O$_3$/MgZnO          | 250            | 0.14              | 1        | 1.72                     | –            | No           | [39] |
|                            | 320            | 2.07              |          |                         |              |              |      |
| GaN nanowire array/graphene| 360            | 6.93 x 10$^{-1}$  | –5       | –                        | 1.23 x 10$^{12}$ | No           | [40] |
|                            | 1540           | 0.11 x 10$^{3}$   |          |                         | 1.88 x 10$^{10}$ |              |      |
| Nanostructure CuO          | 250            | 10                | 20       | 0.18                     | 4 x 10$^{8}$  | No           | [41] |
|                            | 900            | 80                |          |                         | 2.24 x 10$^{11}$ |              |      |
| Nanoporous Ga$_2$O$_3$/GaN | 254            | 43.9              | 0        | 0.04                     | 2.7 x 10$^{11}$ | Yes          | This work |
|                            | 365            | 35.8              |          |                         | 2.17 x 10$^{11}$ |              |      |
and hole diffusion currents. When a reverse bias is applied to the Ga2O3/GaN heterojunction, the built-in potential barriers are increased, and the diffusion of majority carriers is suppressed. The minority carriers (including photogenerated minority carriers) are swept to the other side of the heterojunction by the built-in electric field and form drift currents (Figure 7d).

Figure 7e,f shows the transportation of the photogenerated carriers in the Ga2O3/GaN heterojunction under different illumination wavelengths. At zero bias, the photogenerated electron–hole pairs in the depletion region were separated by the built-in electric field. Under the 254 nm illumination (Figure 7e), the photogenerated holes in Ga2O3 flew to the GaN side, leading to a current in the same direction from n-Ga2O3 to p-GaN). Second, the decrease in the barrier height for the carrier transportation and the increase in the hole concentration gradient between GaN and Ga2O3 greatly enhanced the diffusion current flowing from GaN to Ga2O3, which contributes to the current polarity reversal under the unilateral high-level injection. Based on the photocurrent polarity, the incident illumination bands can be distinguished by the PDs.

3. Conclusion

In conclusion, we designed and fabricated self-powered spectrally distinctive UV PDs based on nanoporous Ga2O3/GaN heterojunctions via the SRE. The effects of nanoporous structures with different porosities on the photoresponse of the PDs were investigated. In the self-powered mode, the nanoporous Ga2O3/GaN film with the highest porosity exhibited the highest responsivity of 43.9 and 35.8 mA W\(^{-1}\) under 254 and 365 nm illumination, respectively. In addition, the nanoporous Ga2O3/GaN PDs showed different current polarities under different illumination wavelengths, which can be used to distinguish the incident light spectrum. This work shows that the nanoporous Ga2O3/GaN heterojunction UV PDs are...
promising for applications in realizing self-powered spectrally distinctive dual-band UV PDs for future photonic systems and applications.

4. Experimental Section

Device Fabrication: GaN thin films were grown on c-plane sapphire substrates by MOCVD (TNSC-MOCVD SR4000), followed by the deposition of high-quality Ga2O3 thin films. The epitaxial layers consisted of 3.5 μm intentionally doped GaN, 300 nm Mg-doped p-GaN (2 × 10^{19} cm^{-3}), and 220 nm unintentionally doped Ga2O3. Subsequently, the Ga2O3 layers were converted into nanoporous films by metal gallium (Ga) SRE. The etching process was also conducted in the MOCVD, where TEGa was used as the Ga precursor. First, the MOCVD reactor temperature was kept at 450 °C. Ga atoms migrated and nucleated at the surface of Ga2O3 leading to Ga droplets in a Volmer–Weber growth mode. Then, the Ga source was turned off, and the reactor temperature was increased to 660 °C. At this high temperature, Ga droplets can react with Ga2O3 to form nanoporous structures.[36] To avoid the depletion of Ga during the SRE process, the Ga source was turned on later when the reaction was initiated. The etching time was 10 min. After the SRE process, the nanoporous films were cleaned in an HF and NH4F mixed solution (BOE, HF:NH4F = 7:1) for 30 min to remove the remaining Ga droplets before the device fabrication. Ti/Au (20/150 nm) bilayer metals were deposited on the nanoporous Ga2O3 film by electron beam evaporation, followed by rapid thermal annealing at 475 °C in N2 ambient to form Ohmic contacts. The underlying p-GaN layer was exposed by selectively etching the nanoporous Ga2O3 films. Then Pd/Pt/Au (30/30 nm) electrodes were deposited on the p-GaN layer by electron beam evaporation and thermally annealed at 575 °C.

Material and Device Characteristics: The Ga2O3/GaN thin films were characterized by an X-ray diffractometer (X’Pert PW3040, X’pert, Bruker D8 Advance) and AFM (Bruker Multimode 8 with ScanAsyst-Air probe). The surface morphologies of the nanoporous films were studied by a scanning electron microscope (SEM). And the reactivity spectra of these films were measured using a spectrophotometer (PerkinElmer LAMBDA 750). The current–voltage and transient response characteristics of the devices were measured by a probe station equipped with Keysight B1505A, commercial 365 nm light-emitting diodes with a light intensity of 30 mW cm⁻², and a shortwave UV lamp tube with a light intensity of 2400 μW cm⁻².

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

current polarity reversal, nanoporous Ga2O3, self-powered UV photodetectors, self-reactive etching, spectrally distinctive

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