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Fermi-Level Splitting-Induced Light-Intensity-Dependent Recombination in Fully Ultra-Wide Bandgap Deep-Ultraviolet Photodetector

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ABSTRACT Ga₂O₃-based heterojunction features the capability of self-driven detection, which is reckoned as a promising candidate for the next-generation deep-ultraviolet (DUV) sensing scenarios. Heterojunction consisting of fully ultra-wide bandgap (UWB) semiconductors would prevent additional response in the near-ultraviolet band. In this work, a β -Ga₂O₃/AlN heterojunction photodetector is constructed and its operating mechanisms are investigated. By measuring its static current-voltage (I - V) and dynamic current-time (I - t) characteristics, the detection performance, including a photo-to-dark current ratio of 8.6×10^5 , a responsivity of 0.41 mA/W, a specific detectivity of 3.4×10^{12} Jones, and an external quantum efficiency of 0.2% were achieved under 0 V bias, indicating that the proposed device realized high-performance self-driven detection. Moreover, this work demonstrated the impact of Fermi-level splitting introduced by the enhanced photo illumination on the carrier recombination and the sensing performance. With the increase of light intensity, Fermi levels are separated and available recombination centers are increased, leading to the enhancement of the recombination process and the variation of detection properties. Consequently, this work highlights the potential of the fully UWB heterojunction and provides further optimization guidelines.

INDEX TERMS β -Ga₂O₃, AlN, heterojunction, photodetector, deep-ultraviolet, recombination.

I. INTRODUCTION

Deep-ultraviolet (DUV) photodetection is less subject to interference from the sunlight since solar irradiation of wavelength shorter than 280 nm rarely reaches the earth's ground. This leads to the important applications of DUV detection in a variety of fields, including scientific research and civilian as well as military equipment [1], [2], [3], [4], [5], [6], [7]. Ultra-wide bandgap semiconductor Ga₂O₃ presents a unique advantage in the DUV sensing application because of its large bandgap of >4.6 eV. Recently, various

Ga₂O₃-based photodetectors including metal-semiconductor-metal (MSM), Schottky barrier diodes (SBDs), field-effect transistors (FETs), and heterojunctions have been proposed and demonstrated with promising detecting efficiency [8], [9], [10], [11], [12], [13], [14], [15], [16].

Ga₂O₃-based heterojunction photodetectors usually composite two materials to form the band alignment and the built-in potential, thereby photo-generated carriers can be driven even without a bias voltage. In addition, the built-in potential prevents carrier transport in the dark

condition and a low dark current can be also achieved. Hence, heterojunction is favored in low-power consumption scenarios. Various semiconductors have been integrated with Ga₂O₃ to construct a heterojunction DUV sensor including ZnO/Ga₂O₃ [17], SnO₂/Ga₂O₃ [18], GaN/Ga₂O₃ [19], and SiC/Ga₂O₃ [20]. These detectors show decent self-driven capability and low dark current thanks to the built-in barrier. However, most reported hetero-materials are of a lower bandgap semiconductor compared to Ga₂O₃. The advantage of ultra-wide bandgap (UWB) such as low response to the near-ultraviolet could be undermined. Therefore, a full UWB heterojunction may sustain the self-powered characteristics as well as the benefits of UWB semiconductors [21], [22].

Meanwhile, lattice mismatch is a crucial issue for heterojunctions as it is the main reason for the interfacial states. A significant number of surface states will act as the recombination centers and exhibit a non-negligible impact on the carrier transport as well as the recombination process. The most thermally stable polymorphs of Ga₂O₃, i.e., monoclinic β-Ga₂O₃ usually exhibit large mismatch to other semiconductors [23]. Therefore, a study of the sensor performance for a β-Ga₂O₃-based heterojunction through exploring the recombination process is critical.

In this work, a fully ultra-wide bandgap self-powered photodetector based on β-Ga₂O₃/AlN heterojunction was demonstrated. The nanofilms of β-Ga₂O₃/AlN were constructed by using the metalorganic chemical vapor deposition (MOCVD) and radio-frequency (RF) sputter technique. Its sensor performance including the static and the dynamic characteristics were measured. Besides, various detection-related figure-of-merits (FOMs) were calculated and studied under different light intensities and biased voltages. Most importantly, the impact of the recombination centers on the photocurrent linearity, dynamic current surge, and response time was evaluated and discussed.

II. EXPERIMENTAL

β-Ga₂O₃ nanofilms were prepared through MOCVD hetero-epitaxy on (0001) sapphire substrates. During epitaxy growth, the chamber temperature and the pressure were ~750 °C and 25 Torr, respectively. Followed by the AlN film deposition using radio-frequency (RF) sputtering. The parameters of RF sputtering are as follows: the RF power was set to 175 W and the deposition time was 2 hours; the substrate temperature is 300 K and the chamber pressure is 0.75 Pa. No further annealing process was conducted. Note that no dopant was introduced during the film deposition. Therefore, high-resistance thin films would be formed. Subsequently, Ti/Au electrodes were formed by using the direct-current (DC) magnetron sputtering and a Cr-metal hard mask. The quality of the β-Ga₂O₃/AlN heterojunction was verified by X-ray diffraction (XRD), scanning electron microscope (SEM), UV-visible (UV-vis) absorbance spectrum, and atomic force microscopy (AFM). A 254 nm UV lamp was adopted as the DUV light source and the light intensity was adjusted by changing the distance from

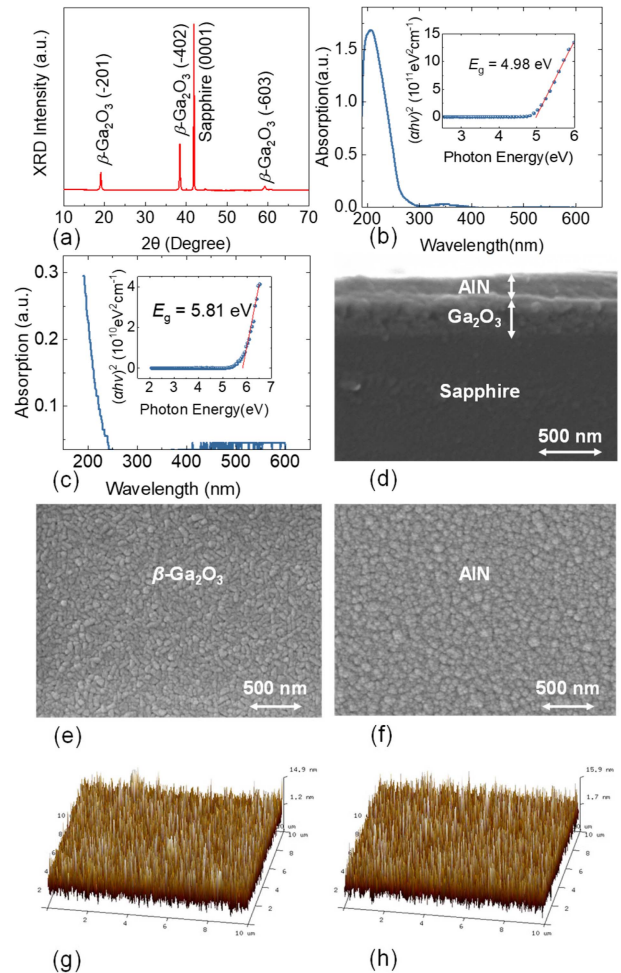


FIGURE 1. (a) XRD θ - 2θ scan of the β -Ga₂O₃/AlN heterojunction. (b) UV-vis absorbance spectrum of the β -Ga₂O₃ film. (c) UV-vis absorbance spectrum of the AlN film. (d) SEM image of the cross-section for the β -Ga₂O₃/AlN heterojunction. (e) and (f) are the surface SEM images of the β -Ga₂O₃ and the AlN, respectively. (g) and (h) are the AFM images of the β -Ga₂O₃ and the AlN, respectively

the light source to the measured sample. A semiconductor analyzer Agilent B1505A with a high-resolution source measurement unit (HR SMU, minimum measured current of 1 fA) was used to record the current-voltage (I - V) and current-time (I - t) curves of the heterojunction photodetector.

III. RESULTS AND DISCUSSION

In this section, we first characterize the crystal quality of the deposited β-Ga₂O₃/AlN films by XRD, SEM, UV-vis, and AFM measurements, as shown in Fig. 1. In Fig. 1(a), the (201), (402) and (603) planes of the β-Ga₂O₃ can be observed. In addition, (0001) sapphire substrate was also verified from the XRD results. No visible peaks of the AlN are seen in Fig. 1(a), suggesting its low crystallinity. Fig. 1(b) shows the UV-vis absorbance spectrum of the β-Ga₂O₃ thin film. It is seen that the β-Ga₂O₃ has marginal absorbance in the visible range but exhibits a rather high absorption in the DUV band, indicating significant

photoresponse shall be presented under DUV radiation. Based on the Tauc relationship, the optical bandgap E_g of the deposited β -Ga₂O₃ can be obtained from the following equation:

$$(\alpha hv)^2 = C(hv - E_g) \quad (1)$$

where α is the absorption coefficient, C is a constant, and hv is the photon energy of the incident light. By extrapolating the $(\alpha hv)^2$ and hv , as seen in the inset figure of Fig. 1(b), the optical bandgap of β -Ga₂O₃ is calculated to be 4.98 eV. In Fig. 1(c), the optical bandgap of the AlN was also determined by the same approach. The cut-off wavelength for the AlN is shorter than the β -Ga₂O₃, which is less than 250 nm, and thus is expected to form a self-powered photodetection without introducing a response to the near-ultraviolet band. Consequently, the measured bandgap of AlN is 5.81 eV. Fig. 1(d) depicts the cross-sectional SEM image of the β -Ga₂O₃/AlN heterojunction. The thickness of the β -Ga₂O₃ and the AlN films are measured to be \sim 260 nm and \sim 180 nm, respectively. Fig. 1(e) and 1(f) display surface SEM images of the β -Ga₂O₃ and the AlN, with grain sizes of approximately 50 nm and 80 nm, respectively. Meanwhile, Fig. 1(g) and 1(h) present their AFM images, indicating roughness of 3.84 nm for β -Ga₂O₃ and 3.95 nm for AlN.

Fig. 2(a) is the device diagram of the fabricated β -Ga₂O₃/AlN heterojunction photodetector. Fig. 2(b) displays a top view of the device observed under a microscope. The effective illuminated area of the device is 2.25 mm². In Fig. 2(c), the I - V characteristics in the dark condition and under various light intensities are compared. With the successful fabrication of the high-resistance films and the formation of built-in potential, the minimum dark current was measured to be \sim 1 fA, which approaches the lowest measurable value. Under the 254 nm DUV illumination, the termination current is improved up to 10^{-8} A. At a 0 V bias voltage, a photocurrent of $>10^{-10}$ A was also measured, suggesting that the self-powered mode is achieved. An open circuit voltage of \sim 0.5 V is obtained due to the built-in potential. In addition, a greater photocurrent was obtained with a higher light intensity, and the linearity of the photocurrent will be discussed later. It can be calculated that the photo-to-dark current ratio $PDCR$ of the detector under 0 V bias is near 10^6 , which is superior to most reported Ga₂O₃ photodetectors [24], [25], [26].

In Fig. 2(d), the photocurrent as a function of the light intensity with various bias voltages is presented. According to a power law [27]: $I_{\text{photo}} \sim kP^\theta$, where I_{photo} is the photocurrent, k is a constant, and P is the light intensity. The linearity can be represented by the value of θ . The slope k of the fitting line is increased with a higher bias voltage due to the enhanced electric field. Meanwhile, $\theta < 1$ was extracted for all bias voltages. Ideally, the θ should be unity providing that a stable carrier recombination process was established. In Rose's model [28], both situations where $\theta < 1$ and $\theta > 1$

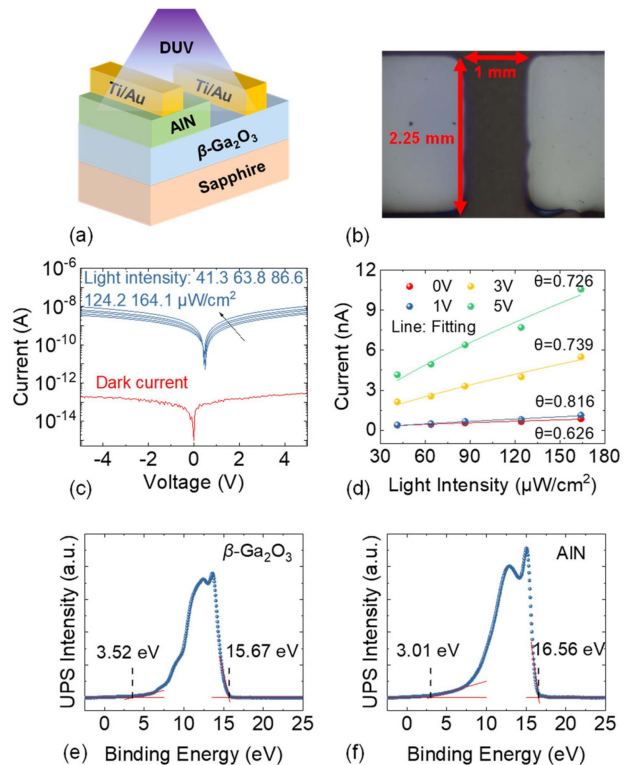


FIGURE 2. (a) Device diagram and (b) top-view micrograph of the β -Ga₂O₃/AlN heterojunction photodetector. (c) Dark current and photocurrent curves with various light intensities. (d) Light-intensity-dependent photocurrent of β -Ga₂O₃/AlN heterojunction. (e) and (f) are the ultraviolet photoelectron spectroscopy of the β -Ga₂O₃ and the AlN, respectively.

were thoroughly discussed and can be well applied in the sub-linearity phenomenon of this work.

Fig. 2(e) and 2(f) present the ultraviolet photoelectron spectroscopy (UPS) for β -Ga₂O₃ and AlN, respectively. The calculated work functions of β -Ga₂O₃ and AlN are 21.22-15.67=5.55 eV and 21.22-16.56=4.66 eV respectively, where 21.22 is the energy value of helium discharge lamp photons. Additionally, the valence band edge values (relative to the Fermi level) for both materials are extracted to be 3.52 eV and 3.01 eV, respectively.

To gain insight into the self-powered operational mechanism and sub-linear photocurrent behavior, Fig. 3(a) and 3(b) depict the energy band diagrams of the β -Ga₂O₃/AlN heterojunction before and under illumination. Based on the measured bandgaps and UPS data of the β -Ga₂O₃ and AlN, a valence band offset of 1.40 eV and a conduction band offset of 2.23 eV were obtained. The β -Ga₂O₃/AlN heterojunction exhibits a typical type II heterojunction. With a DUV illumination, photo-generated electrons are excited from the valence band to the conduction band. The carriers are then transported by the built-in electric field and the self-driven photocurrent is formed.

Due to the lattice mismatch between the AlN and the β -Ga₂O₃, the interface states as well as the bulk defects are expected to behave as the main recombination centers. These

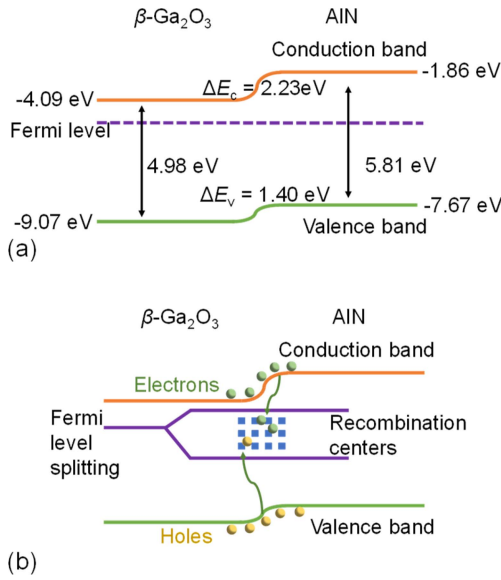


FIGURE 3. Energy band diagram of the $\beta\text{-Ga}_2\text{O}_3/\text{AlN}$ heterojunction (a) before illumination and (b) under illumination.

centers are critical for the carrier transport process. For a better understanding, Fig. 3(b) also illustrates the recombination centers. Without DUV illumination, a thermal equilibrium was achieved and the photodetector has a unified Fermi level. Under the DUV light illumination, carriers are generated and the quasi-Fermi-level for electrons (QFLE) and quasi-Fermi-level for holes (QFLH) are separated due to the nonequilibrium carriers' generation. With the enhancement of the light intensity, more carriers are generated and thus the quasi-Fermi-levels (QFLs) are more separated, leading to the increase of unoccupied recombination centers. Consequently, the recombination rate is improved and the concentration of available carriers that can form the photocurrent is reduced, resulting in the deviation of the intensity-photocurrent curve from linearity.

It is well known that the external voltage can also lead to the nonequilibrium carrier injection. In other words, as the biased voltage increases, QFLs would be separated as well and the θ should be further reduced. However, the θ exhibits no correlation with the applied voltage, which can be found in Fig. 2(d). This implies that the QFLs' separation is mainly determined by the incident DUV light rather than the applied voltage.

We then examine the detection figure-of-merits (FOMs) including responsivity R , photo-to-dark current ratio $PDCR$, detectivity D^* , and external quantum efficiency EQE , which are described in equation (2)–(5):

$$R = \frac{(I_{photo} - I_{dark})}{P \cdot S} \quad (2)$$

$$PDCR = \frac{I_{photo}}{I_{dark}} \quad (3)$$

$$D^* = \frac{R}{\sqrt{2q \frac{I_{dark}}{S}}} \quad (4)$$

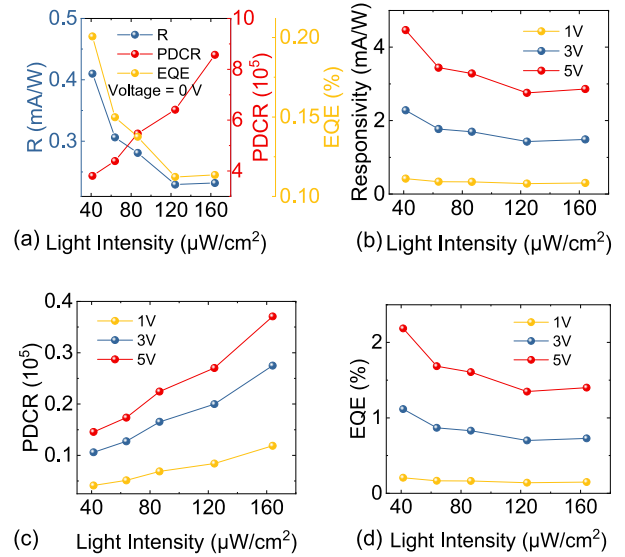


FIGURE 4. (a) Responsivity R , photo-to-dark current ratio $PDCR$, and external quantum efficiency EQE at 0 V bias. (b)–(d) are R , $PDCR$, and EQE at various applied voltages, respectively.

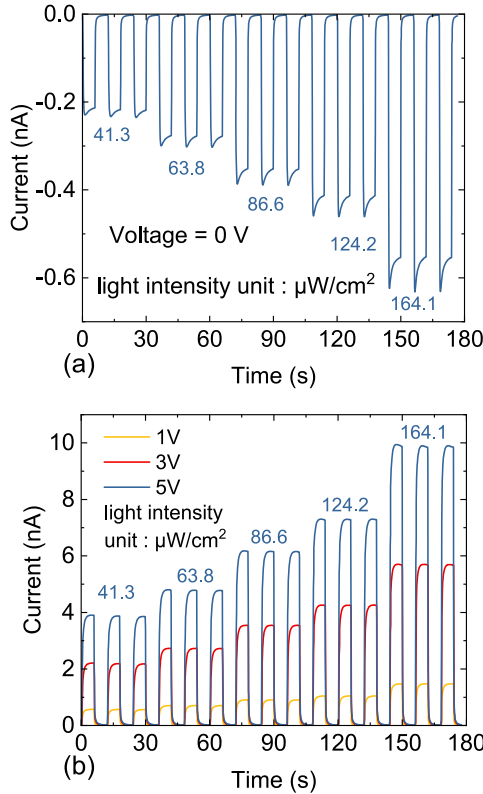
$$EQE = \frac{Rhc}{q\lambda} \quad (5)$$

where I_{photo} and I_{dark} are the photocurrent and dark current, respectively, P is the light intensity, S is the effective illuminated area, q is the elementary charge, h is Planck's constant, c is the light velocity, and λ is the light wavelength. In Fig. 4(a), the R , $PDCR$, and the EQE of the $\beta\text{-Ga}_2\text{O}_3/\text{AlN}$ sensor at 0 V bias are extracted. At a light intensity of $41.3 \mu\text{W}/\text{cm}^2$, a maximum responsivity of $0.41 \text{ mA}/\text{W}$ is obtained. In addition, a $PDCR$ of 8.6×10^5 and an EQE of 0.2% is achieved. The detectivity of 3.4×10^{12} Jones was obtained at a light intensity of $41.3 \mu\text{W}/\text{cm}^2$. Meanwhile, all the FOMs are degraded at a higher light intensity. Photodetection FOMs fundamentally depend on the competition between carrier transport and recombination processes. In detail, higher transport efficiency leads to the improvement of FOMs mentioned above. The interface recombination states and bulk defect states in $\beta\text{-Ga}_2\text{O}_3$ and AlN can lead to the re-capture and recombination of photogenerated charge carriers, impeding their efficient transport to the electrodes. The improvement of interface and bulk quality is beneficial to the FOMs of a heterojunction photodetector [29], [30], [31], [32], [33]. Nevertheless, Table 1 provides a comparison of the self-powered performance between various Ga_2O_3 heterojunction photodetectors [34], [35], [36], [37], [38], [39]. The proposed $\beta\text{-Ga}_2\text{O}_3/\text{AlN}$ heterojunction photodetector exhibits excellent detection capabilities.

The carrier transport efficiency can also be improved by applying an external voltage. Fig. 4(b)–(d) shows the responsivity, $PDCR$, and EQE with various bias voltages. It is seen that the FOMs are improved with a greater applied voltage due to enhanced carrier transport. At an external voltage of 5 V and a light intensity of

TABLE 1. Comparison of self-powered performance for Ga₂O₃ heterojunction photodetectors.

PD	PDCR	R (mA/W)	D* (Jones)	Rise Time (ms)	Decay Time (ms)	Ref.
CuCrO ₂ /Ga ₂ O ₃	3.5×10 ⁴	0.1	4.7×10 ¹¹	350/1850	60	[34]
Diamond/β-Ga ₂ O ₃	37.0	0.2	7.0×10 ⁹	N/A	N/A	[35]
ZnO/β-Ga ₂ O ₃	1.0×10 ⁴	9.7	6.3×10 ¹²	0.1	0.9	[36]
CuI/β-Ga ₂ O ₃	4.0×10 ³	8.5	6.3×10 ¹²	97.8	28.9	[37]
NiO/Ga ₂ O ₃	1.0×10 ²	0.3	1.8×10 ⁸	12/41	8/75	[38]
MoS ₂ /β-Ga ₂ O ₃	1.3×10 ⁴	2.1	1.2×10 ¹¹	N/A	N/A	[39]
AlN/β-Ga ₂ O ₃	8.6×10 ⁵	0.4	3.4×10 ¹²	129	136	this work


FIGURE 5. Temporal response of the β-Ga₂O₃/AlN photodetector with various light intensities at (a) 0 V and (b) 1, 3, 5 V.

41.3 $\mu\text{W}/\text{cm}^2$, the photodetector exhibits R , $PDCR$, and EQE of 4.5 mA/W, 1.5×10^4 , and 2.2%, respectively, suggesting that the performance can be further improved at the expense of power consumption. In addition, the degradation caused by the improved light intensity is much lower than that of 0 V. This is because the carriers are drained rapidly by the enhanced electric field and thus fewer carriers would be recombined.

Furthermore, the temporal response of the β-Ga₂O₃/AlN photodetector is studied to determine its transient characteristics, as shown in Fig. 5. During the measurement, a duty time of 12 s and a duty cycle of 50% were used. Fig. 5(a) presents the I - t characteristics without applied voltage and Fig. 5(b) is that with applied voltage. It is observed that the dark current is negative, and its magnitude is enhanced with greater light intensity, as shown in Fig. 5(a). For applied voltages of 1, 3, and 5 V, positive photocurrent can be observed and the same trend can be found when

the light intensity increases. Meanwhile, for self-powered operation, a visible photocurrent surge was observed at the beginning of the DUV illumination stage. With an enhanced light intensity, the surge peak was also enhanced yet vanished when an externally applied voltage was introduced.

This phenomenon can be attributed to the carrier generation/recombination relaxation process. At the start of DUV illumination, carriers are generated within the β-Ga₂O₃/AlN heterojunction and are driven to form the photocurrent at 0 V bias. Compared to a positive-voltage situation, the built-in electric field is relatively low. The insufficient built-in electric field results in the low-efficiency carrier transport and more photo-generated carriers will be recombined rather than transported. This relaxation process is the main reason for the photocurrent spike in Fig. 5(a). Providing sufficient time, the recombination/generation reaches a stable state and thus a steady photocurrent is obtained.

Another fact is that an enhanced current peak was observed when the light intensity was increased. It is ready to conclude that QFLs are much separated and the recombination process is improved in that case, leading to a more significant photocurrent overshoot. Once the external electric field was applied, the carrier transport was promoted and fewer carriers would be recombined. Therefore, the relaxation duration is almost eliminated and no significant photocurrent peak can be observed, as shown in Fig. 5(b).

In a MSM photodetector, the current peak is hardly observed since a bias voltage is applied. With a heterojunction photodetector, to pursue a self-powered operation, the device is designed to operate under a zero-bias condition. Therefore, to obtain a surge-free transient characteristic, a band alignment that features a larger built-in potential and a higher electric field is preferred. This means that the optimization of material selection and doping concentration could be beneficial.

We then further examine the transient characteristics of the heterojunction photodetector, as shown in Fig. 6(a)-(e). The response time can be extracted by $I = I_0 + Ae^{-\frac{t}{\tau}}$ [27], where I_0 is the stable state I_{photo} , A is a constant, t is the time, and τ is a time constant. The rise and decay time (τ_r and τ_d) of the β-Ga₂O₃/AlN sensor without external voltage are also displayed in Fig. 6(a)-(e). The rise time and the decay time at 41.3 $\mu\text{W}/\text{cm}^2$ are 207 ms and 159 ms, respectively. Both the rise time and the decay time are decreased with enhanced light intensity.

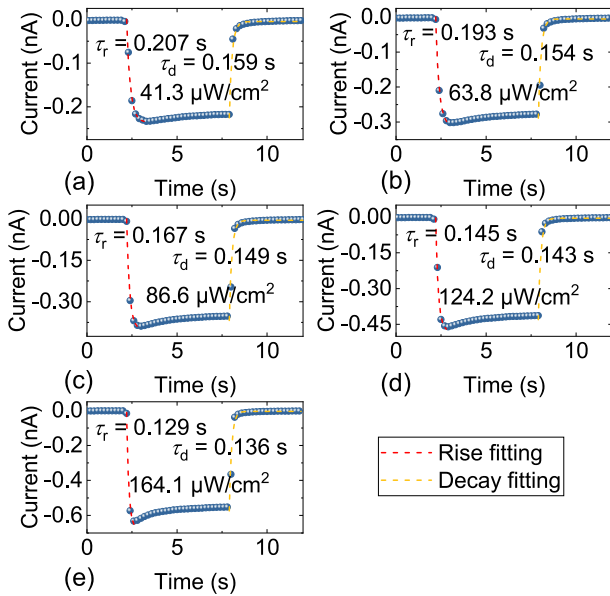


FIGURE 6. (a)–(e) Single on/off cycle of the β -Ga₂O₃/AlN heterojunction photodetector under different light intensities. The rise time and decay time are extracted and plotted in each figure.

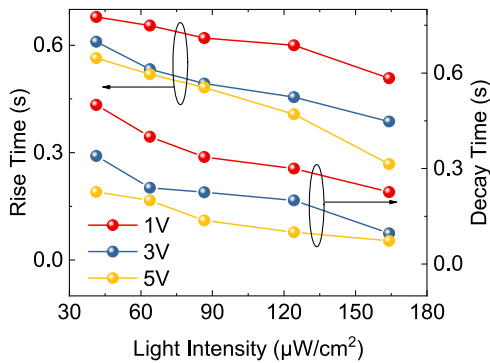


FIGURE 7. Extracted rise time and decay time of the photodetector as a function of the light intensity with applied voltages.

The response time, especially for the decay time, is strongly correlated to the SRH recombination rate. In other words, a higher recombination rate will result in a faster response time. As we mentioned above, a wider separation of the QFLs is formed when the light intensity increases. The origin of the $\theta < 1$ and the decreased response time are essentially the same, that is, more available recombination centers and improved recombination rate. Fig. 7 also shows the response time as a function of the light intensity with applied voltages. The same trend that both rise time and decay time are decreased with a higher light intensity can be found.

Although the recombination centers are beneficial to the response time, one should further improve the material quality and reduce the defects since they are unintentionally introduced and are harmful to the detection linearity. As the maturity of β -Ga₂O₃/AlN crystal growth and heteroepitaxy is under rapid development, Ga₂O₃-based heterojunction with

a greater sensing performance will be realized in the near future.

IV. CONCLUSION

To conclude, in this work, we propose a β -Ga₂O₃/AlN heterojunction photodetector, and its operating principles are investigated. The I - V and I - t characteristics are measured, and the detection performance, including a photo-to-dark current ratio of 8.6×10^5 , a responsivity of 0.41 mA/W, a specific detectivity of 3.4×10^{12} Jones, and an external quantum efficiency of 0.2% were achieved under 0 V bias. More importantly, by exploring the synergetic effect between the carrier transport and recombination process, this work demonstrates the Fermi-level splitting and its impact on the detection figure-of-merits. In all, this work shows a high-performance self-powered DUV photodetection and lays the foundation for further development of fully UWB photodetectors.

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