Single-crystalline all-oxide α - γ - β heterostructures for deep-ultraviolet photodetection

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ABSTRACT:

Recent advancements in gallium oxide (Ga_2O_3)-based heterostructures have allowed optoelectronic devices to be used extensively in the fields of power electronics and deep-ultraviolet (DUV) photodetection. While most previous research has involved realizing single-crystalline Ga_2O_3 layers on native substrates for high conductivity and visible-light transparency, presented and investigated herein is a single-crystalline $\beta\text{-Ga}_2\text{O}_3$ layer grown on an $\alpha\text{-Al}_2\text{O}_3$ substrate through an interfacial $\gamma\text{-In}_2\text{O}_3$ layer. The single-crystalline transparent-conductive-oxide layer made of wafer-scalable $\gamma\text{-In}_2\text{O}_3$ provides the high carrier transport, visible-light transparency, and antioxidation properties that are critical for realizing vertically oriented heterostructures for transparent-oxide photonic platforms. Physical characterization based on X-ray diffraction and high-resolution transmission electron microscopy imaging confirms the single-crystalline nature of the grown films and the crystallographic orientation relationships among the monoclinic $\beta\text{-Ga}_2\text{O}_3$, cubic $\gamma\text{-In}_2\text{O}_3$, and trigonal $\alpha\text{-Al}_2\text{O}_3$, while the elemental composition and sharp interfaces across the heterostructure are confirmed using Rutherford backscattering spectrometry. Furthermore, the energy-band offsets are determined using X-ray photoelectron spectroscopy at the $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3$ interface, elucidating a type-II heterojunction with conduction- and valence-band offsets of 0.16 and 1.38 eV, respectively. Based on the single-crystalline $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ all-oxide heterostructure, a vertically oriented DUV photodetector is fabricated that exhibits a high photoresponsivity of 94.3 A/W, an external quantum efficiency of $4.6 \times 10^4\%$ and a specific detectivity of 3.09×10^{12} Jones at 250 nm. The present demonstration lays a strong foundation for and paves the way to future all-oxide-based transparent photonic platforms.

1. INTRODUCTION

The emergence of highly integrated silicon-based photonic platforms has led to extensive applications in the semiconductor and telecommunications industries. However, despite allowing large-volume manufacturing at relatively low cost, the energy bandgap located near 1.14 eV hinders the integration of devices requiring optical transparency in the ultraviolet (UV) and visible regimes, such as visible-light and deep-UV (DUV) photodetectors,¹⁻⁴ group-III-V-based light-emitting diodes,⁵⁻⁸ solar cells,⁹ electro-absorption modulators,¹⁰⁻¹² and transparent thin-film

transistors.^{13–15} Despite significant efforts to realize the heterogeneous integration of the aforementioned devices on silicon-based platforms,^{16–19} challenges related to high defect densities, optical coupling, wafer bonding, and substrate removal remain critical and can lead to consequential overhead production costs.²⁰ Recently, oxide-based photonic platforms relying on aluminum oxide (Al_2O_3), with an ultra-large optical bandgap of up to ~ 7.6 eV,²¹ have attracted considerable attention as paradigm-shifting platforms for various transparent optoelectronic devices.^{22–25} The realization of low-loss Al_2O_3 waveguides, with an extended wavelength of up to 220 nm and substantially lower transmission losses compared to silicon nitride (Si_3N_4)-based waveguides,^{22,26} is highly encouraging and paves the way for an integrated transparent oxide-based photonic platform across the visible wavelength region. Undeniably, such a platform would also allow the direct growth of conventional group-III-V-based light-emitting devices without the need for tedious photoelectrochemical etching for substrate removal and wafer bonding as in the case of heterogeneous integration on silicon-based platforms.^{27,28} The eventual realization of a transparent all-oxide-based photonic platform would find promising applications in next-generation augmented-reality displays, smart goggles, smart windows, head-up displays, and other see-through devices.^{29–31} This would be the case particularly where large-bandgap DUV photodetectors could be integrated as monitoring sensors for harmful ultraviolet-C (UVC) radiation (i.e., wavelengths of 100–280 nm) coming from flames and other man-made sources (e.g., mercury lamps, welding torches, disinfection lamps), while maintaining high transparency across the visible wavelength region.

With a large optical bandgap of up to ~ 5 eV, gallium oxide (Ga_2O_3) has enabled emerging and attractive forefronts for next-generation oxide-based high-voltage power rectifiers,³² field-effect transistors,^{33,34} and DUV photodetectors^{1,25}. This is largely because Ga_2O_3 has the inherent advantages of (i) high breakdown fields of ~ 8 MV/cm, (ii) electron mobilities of ~ 150 $\text{cm}^2/\text{V}\cdot\text{s}$, and (iii) ultra-high-responsivity photodetection due to deep trap states formed by oxygen vacancies.^{35–38} However, despite Ga_2O_3 being suited for transparent oxide-based photonic platforms and previous work showing heteroepitaxial growth of Ga_2O_3 on an Al_2O_3 platform,^{1,39–41} a high crystalline quality and conductive intermediate layer are necessary to ensure high carrier mobility, low Schottky barrier, and sufficiently high optical transparency in the visible wavelength region. Although using interfacial metal layers or other two-dimensional materials (e.g., transition metal dichalcogenides,⁴² hexagonal boron nitride,⁴³ graphene^{35,44}) could provide the required

electrical and optical properties, those methods rely on chemical vapor deposition (CVD), thermal evaporation, and sputtering and typically result in polycrystalline structures that are not ideal for the subsequent growth of high-quality crystal structures. However, single-crystalline indium oxide (In_2O_3), an intrinsically n-type transparent conducting oxide (TCO) layer with an optical bandgap of ~ 3.7 eV,^{45–47} has the potential to achieve the aforementioned goals and emerge as the material of choice as a conducting interfacial layer between the Ga_2O_3 and Al_2O_3 platforms. Recently, Sun et al.⁴⁶ demonstrated type-I band alignment with narrow conduction-band offset (CBO) and valence-band offset (VBO) of 0.35 and 0.45 eV, respectively, at the polycrystalline $\beta\text{-Ga}_2\text{O}_3/\text{In}_2\text{O}_3$ interface, elucidating the potential of the above heterostructure for reducing the barrier height and enhancing carrier transport in visible-transparent oxide platforms. Similarly, Zhi et al.⁴⁷ demonstrated an even lower CBO of 0.07 eV in the type-I heterostructure for a pulsed laser deposition (PLD)-grown polycrystalline $\text{Ga}_2\text{O}_3/\text{In}_2\text{O}_3$ heterostructure. However, although previous studies have investigated the band alignment^{46–48} and electronic properties⁴⁹ of the aforementioned heterostructures, the related crystallinity properties of the as-grown layer were largely limited to amorphous or polycrystalline,^{46,50} and so those studies shed no light on the high-quality single-crystalline $\beta\text{-Ga}_2\text{O}_3/\text{In}_2\text{O}_3$ heterostructures that are critical for realizing various high-performing oxide-based optoelectronics devices or platforms. Moreover, optoelectronics applications based on $\beta\text{-Ga}_2\text{O}_3/\text{In}_2\text{O}_3$ heterostructures grown on Al_2O_3 -based platforms are yet to be demonstrated.

Herein, we report on a PLD-grown group-III-oxide $\alpha\text{-}\gamma\text{-}\beta$ heterostructure comprising single-crystalline and thermodynamically stable $\gamma\text{-In}_2\text{O}_3$ and $\beta\text{-Ga}_2\text{O}_3$ layers grown sequentially on sapphire, as well as a DUV photodetector based on this all-oxide platform. The successful and direct growth of high-quality intermediate TCO comprising a single-crystalline $\gamma\text{-In}_2\text{O}_3$ layer with inherent antioxidation properties is a critical step to enable high carrier transport and the subsequent growth of a functional single-crystalline absorptive oxide layer integrated in the platform. Physical characterization—namely X-ray diffraction (XRD), Rutherford backscattering spectrometry (RBS), high-resolution transmission electron microscopy (HR-TEM), and X-ray photoelectron spectroscopy (XPS)—is used to further our understanding of the fundamental properties of the as-grown heterostructure interfaces. In particular, the combination of XRD and HR-TEM imaging confirms the high-crystallinity of and orientation relationships among the $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ stacked layers. The chemical composition of the proposed stack is

confirmed through elemental mapping based on scanning-TEM high-angle annular dark-field (STEM-HAADF) imaging and RBS characterization. Also, the interfacial properties related to the energy-band offsets in the β -Ga₂O₃/ γ -In₂O₃ heterostructure are analyzed using XPS and are found to have a CBO and VBO of 0.16 eV and 1.38 eV, respectively, with type-II alignment. To demonstrate an optoelectronic device based on the single-crystalline β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterostructure, a DUV photodetector is fabricated and its photoelectrical performance is characterized in terms of its spectral responsivity (R_λ), external quantum efficiency (EQE), specific detectivity (D^*), and time response characteristics. Our demonstration based on the single-crystalline β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterostructure paves the way for vertically structured optoelectronic devices in the paradigm-shifting all-oxide-based photonic platforms.

2. EXPERIMENTAL METHODS

2.1 Thin Film Growth

The 460- μ m-thick α -Al₂O₃ substrate (Figure 1a) was obtained commercially from Cryscore Optoelectronic Ltd. After standard cleaning with acetone and isopropyl alcohol, the substrate was placed in a PLD chamber that was pumped down to a base pressure of 10^{-8} Torr. The conductive γ -In₂O₃ layer was grown atop the α -Al₂O₃ substrate at an oxygen (O₂) partial pressure of 5 mTorr and a substrate temperature of 500°C. A KrF excimer laser was used, working at 248 nm, a frequency of 1 Hz, an energy of 300 mJ/pulse, and a fluence of 2 J/cm². The very low growth rate of 1 Hz was used to obtain single-crystalline γ -In₂O₃ by overcoming the large lattice mismatch between α -Al₂O₃ and γ -In₂O₃. In total, 3000 pulses were used to grow the 180-nm-thick γ -In₂O₃ layer at a deposition rate of ~ 0.6 Å/pulse (Figure 1b). After growing the γ -In₂O₃ layer, the O₂ pressure was set at 100 mTorr to prevent γ -In₂O₃ evaporation, and the substrate temperature was increased to 800°C at a rate of 20°C/min. When the substrate temperature reached 800°C, β -Ga₂O₃ was deposited at an O₂ partial pressure of 5 mTorr. Similarly, the laser was optimized to have a pulse frequency of 5 Hz, an energy of 300 mJ/pulse, and a fluence of 2 J/cm². In total, 30 000 pulses were used to grow the 350-nm-thick β -Ga₂O₃ at a deposition rate of ~ 0.12 Å/pulse (Figure 1c). For the deposition of the γ -In₂O₃ and β -Ga₂O₃ layers, the target-to-substrate distance was set at 95 and 80 mm, respectively. The thickness of the γ -In₂O₃ layer and that of the β -Ga₂O₃ layer were determined via TEM analysis.

2.2 X-ray Crystallography

The crystallographic properties of the group-III-oxide α - γ - β heterostructures (i.e., β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃) were examined using Bruker D8 ADVANCE (out-of-plane) and D8 DISCOVER (in-plane) X-ray diffractometers using Cu K α radiation ($\lambda = 1.5405 \text{ \AA}$).

2.3 Rutherford Backscattering Spectrometry (RBS)

To determine the structural and material properties of the grown α - γ - β heterostructures, the β -Ga₂O₃/ γ -In₂O₃ (150 nm/80 nm) bilayer grown on the α -Al₂O₃ substrate was examined using a high-resolution RBS system (HRBS-V500; Kobe Steel Ltd., Japan). Prior to the measurement, the sample was pre-coated with Ir metal to prevent charge accumulation on the surface. A detection angle of 107.5° and a 400-keV beam of He⁺ ions were used throughout the measurement.

2.4 High-resolution Transmission Electron Microscopy (HR-TEM)

The HR-TEM lamella of the α - γ - β heterostructures were prepared using an FEI Helios G4 dual-beam focused-ion-beam scanning-electron-microscope system equipped with an OmniProbe and a gallium ion source. The HR-TEM images and fast Fourier transform (FFT) patterns of the β -Ga₂O₃/ γ -In₂O₃ and γ -In₂O₃/ α -Al₂O₃ interfaces were acquired using an HR-TEM system (Titan ST microscope; FEI, USA) operating at 300 keV, and image analysis was performed using a Gatan DigitalMicrograph®. The acquired FFT patterns of each layer extracted from the HR-TEM images were matched with the corresponding FFT patterns created using CrystalMaker®. The crystal models for β -Ga₂O₃, γ -In₂O₃, and α -Al₂O₃ were created using VESTA (Visualization for Electronic and Structural Analysis).

2.5 X-ray Photoelectron Spectroscopy

To determine the energy-band offset at the β -Ga₂O₃/ γ -In₂O₃ heterojunction interface, an XPS system (Axis Supra DLD; Kratos, UK) with a monochromatic source of Al K α radiation (1486.6 eV) operating at 150 W was used. To avoid differential charging, the sample was mounted in floating mode and the XPS spectra were acquired using a fixed analyzer pass energy of 20 eV. The C 1s binding of adventitious carbon contamination, taken at 284.8 eV, was used as the reference for all other binding energies.

2.6 Device Fabrication

After the epitaxial growth of the β -Ga₂O₃ and γ -In₂O₃ thin films (Figure 1c), a mesa-type structure was formed using an inductively coupled plasma reactive ion etcher operating at a chamber pressure of 5 mTorr with 20 sccm of Ar, 5 sccm of Cl₂, an RF power of 700 W, and a DC voltage of 300 V for 7 min after a photolithography step (see Figure 1d). Metal contacts were then deposited [Au/Ti (180 nm/20 nm)] on the γ -In₂O₃ and β -Ga₂O₃ films using magnetron sputter deposition and were subsequently annealed at 600°C for 1 min in an Ar ambient environment to form ohmic contacts. The final structures were formed with six parallel interdigitated Au/Ti metal fingers with 50- μ m spacing (see Figure 1e).

2.7 Device Measurement

The photoelectrical performance of the photodetector was tested under DUV illumination using a 500-W broadband mercury–xenon arc lamp (66142; Newport, USA). Before illuminating the photodetector, the broadband light passed through a monochromator (Cornerstone 260; Oriel, USA) fitted with a diffraction grating (74060; Newport). The light intensity was pre-calibrated using an Si-based photodetector and controlled using a set of neutral-density filters. The I – V characteristics were extracted using four-terminal Kelvin probes and a semiconductor parameter analyzer (4156C; Agilent Technologies, USA).

Figure 1. Fabrication flow for vertically oriented β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ photodetector: (a) bulk α -Al₂O₃ substrate cleaned and prepared; (b) pulsed laser deposition (PLD) of 180-nm-thick γ -In₂O₃ layer; (c) subsequent PLD of 350-nm-thick β -Ga₂O₃ layer; (d) mesa formation with device area of 0.25 mm² via photolithography and dry etching; (e) metal-contact deposition [Au/Ti (180 nm/20 nm)] using magnetron sputtering, followed by annealing for ohmic contact formation on β -Ga₂O₃ and γ -In₂O₃ layers.

3. RESULTS AND DISCUSSION

3.1 Structural Characterization

Figure 2a shows the out-of-plane XRD measurements for the α - γ - β heterostructures comprising the β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ stack. The XRD peaks corresponding to the β -Ga₂O₃ ($\bar{2}01$), γ -In₂O₃ (222), β -Ga₂O₃ ($\bar{4}02$), α -Al₂O₃ (006), β -Ga₂O₃ ($\bar{6}03$), and γ -In₂O₃ (444) reflections identified at $2\theta \approx 19.26^\circ$, 30.67° , 38.68° , 42.01° , 59.34° , and 63.35° , respectively, match those from the XRD simulation using CrystalDiffract. Note also that the small XRD peak that appears around 27.612° is attributed to the tetragonal-indium (100) plane that could have appeared because of the low O₂ partial pressure (i.e., 5 mTorr) used during the PLD growth process, whereby the indium adatoms could not bind with the oxygen adatoms in the crystal. However, its intensity is more than one to two orders of magnitude lower than that of the primary γ -In₂O₃ (222) or γ -In₂O₃ (444) peak, and so it does not affect the crystallinity of the as-grown γ -In₂O₃ layer. The rocking curve (RC) measurements of the β -Ga₂O₃ ($\bar{2}01$) and γ -In₂O₃ (222) diffractions are shown in Figures 2b and 2c, respectively. By fitting the diffraction profiles using a Gaussian distribution, the full width at half maximum (FWHM) values were calculated to be approximately $2.639 \pm 0.016^\circ$ and $0.995 \pm 0.003^\circ$ for β -Ga₂O₃ ($\bar{2}01$) and γ -In₂O₃ (222), respectively, manifesting high-quality oxide-based crystal structures grown on an α -Al₂O₃ substrate for the intended all-oxide based platform. Based on the out-of-plane XRD measurements, the crystallographic relationship of the α - γ - β heterostructures can be identified as β -Ga₂O₃ ($\bar{2}01$) $\parallel$ γ -In₂O₃ (222) $\parallel$ α -Al₂O₃ (006).

Correspondingly, the plane spacings ($d_{(hkl)}$) for monoclinic β -Ga₂O₃ ($\bar{2}01$), cubic γ -In₂O₃ (222), and trigonal α -Al₂O₃ (006) were calculated using Bragg's law (i.e., $2d_{(hkl)} \sin \theta = n\lambda$) and the following interplanar spacing formulas::

$$\frac{1}{d_{\beta\text{-Ga}_2\text{O}_3}^2} = \frac{1}{\sin^2\beta} \left(\frac{h^2}{a^2} + \frac{k^2 \sin^2\beta}{b^2} + \frac{l^2}{c^2} - \frac{2hl \cos\beta}{ac} \right) \quad (1)$$

$$\frac{1}{d_{\gamma\text{-In}_2\text{O}_3}^2} = \frac{h^2 + k^2 + l^2}{a^2} \quad (2)$$

$$\frac{1}{d_{\alpha\text{-Al}_2\text{O}_3}^2} = \frac{4}{3} \left(\frac{h^2 + hk + k^2}{a^2} \right) + \frac{l^2}{c^2} \quad (3)$$

where a , b , and c are the lattice parameters along the different axes, h , k , and l are the Miller indices, and β is the angle between the a and c axes. The Miller indices were determined from the first observed spot above the center of each FFT pattern. Based on the above, the plane spacings for $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}01$), $\gamma\text{-In}_2\text{O}_3$ (222), and $\alpha\text{-Al}_2\text{O}_3$ (006) were determined to be 4.6857, 2.9453, and 2.1651 Å, respectively.

The in-plane XRD measurements, where the crystal planes are aligned azimuthally across the heterostructure, are shown in Figure 2d. The φ scan shows six corresponding peaks with 60° interval on the $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}21$), $\gamma\text{-In}_2\text{O}_3$ (004), and $\alpha\text{-Al}_2\text{O}_3$ (103) planes. Therefore, the orientation relationship identified based on the in-plane XRD measurements is $\beta\text{-Ga}_2\text{O}_3$ (020) $\parallel$ $\gamma\text{-In}_2\text{O}_3$ ($10\bar{1}$) $\parallel$ $\alpha\text{-Al}_2\text{O}_3$ (100).

The RC measurements for $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}21$) and $\gamma\text{-In}_2\text{O}_3$ (004) are shown in Figures 2e and 2f, respectively. The FWHM values extracted from fitting a Gaussian distribution were identified to be $4.627 \pm 0.031^\circ$ for $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}21$) and $4.237 \pm 0.035^\circ$ for $\gamma\text{-In}_2\text{O}_3$ (004).

Figure 2. (a) Out-of-plane X-ray diffraction (XRD) measurements for β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterostructure. Corresponding rocking curve (RC) and full width at half maximum (FWHM) measurements for (b) β -Ga₂O₃ ($\bar{2}01$) and (c) γ -In₂O₃ (222) diffractions. (d) In-plane XRD measurements for β -Ga₂O₃ and γ -In₂O₃ grown on α -Al₂O₃ substrate. Corresponding RC and FWHM measurements for (e) β -Ga₂O₃ ($\bar{2}21$) and (f) γ -In₂O₃ (004) diffractions.

3.2 Electron Microscopy Analysis

Figures 3a and 3b show HR-TEM images of the β -Ga₂O₃/ γ -In₂O₃ and γ -In₂O₃/ α -Al₂O₃ interfaces, respectively, confirming that they are high-crystallinity interfaces with a sharp layer transition. The corresponding FFT patterns acquired for each layer located closed to the interfaces are shown in Figures 3c and 3d, elucidating the symmetry of the lattice fringes on the HR-TEM images. To distinguish among the diffraction spots observed in the FFT images, crystallographic analysis was performed using the CrystalMaker software and matched with the acquired FFT images. The orientation relationships for β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ as resolved from the FFT analysis are β -Ga₂O₃ ($\bar{2}01$) $\parallel$ γ -In₂O₃ (222) $\parallel$ α -Al₂O₃ (003) and β -Ga₂O₃ (020) $\parallel$ γ -In₂O₃ ($\bar{2}02$) $\parallel$ α -Al₂O₃ (300).

The orientation relationships acquired from the FFT analysis match the out-of-plane and in-plane XRD, as presented earlier in Figure 2. Also, the plane spacings for the monoclinic β -Ga₂O₃

($\bar{2}01$), cubic $\gamma\text{-In}_2\text{O}_3$ (222), and trigonal $\alpha\text{-Al}_2\text{O}_3$ (003) layers obtained from the FFT patterns were determined to be 4.7619, 2.8985, and 2.1739 Å, respectively, which agree closely with the 2θ scan in the out-of-plane XRD measurement.

Figure 4b shows the elemental mapping acquired based on STEM-HAADF in the selected region of the $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ stack (see Figure 4a), revealing the layer compositions and thicknesses and the minimal thermally induced interdiffusion region. Figures 4c–4e show the VESTA crystal models for the various layer structures, where the colored slice planes represent $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}01$), $\gamma\text{-In}_2\text{O}_3$ (222), and $\alpha\text{-Al}_2\text{O}_3$ (006), respectively. Each slice plane shows the hexagonal planar structure that enables monoclinic and cubic crystals to be grown epitaxially on a trigonal crystal. However, while $\beta\text{-Ga}_2\text{O}_3$ and $\gamma\text{-In}_2\text{O}_3$ have monoclinic and cubic crystal structures, respectively, the $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}01$) and $\gamma\text{-In}_2\text{O}_3$ (222) planes have a hexagonal structure. The lattice constant of $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}01$) was found to be 2.95 Å whereas that of $\gamma\text{-In}_2\text{O}_3$ (222) was 3.35 Å, thereby resulting in a lattice mismatch between the $\beta\text{-Ga}_2\text{O}_3$ ($\bar{2}01$) and $\gamma\text{-In}_2\text{O}_3$ (222) layers of 13.55%. Meanwhile, the trigonal $\alpha\text{-Al}_2\text{O}_3$ (006) has a hexagonal structure with a lattice constant of 2.74 Å, and the lattice mismatch between $\gamma\text{-In}_2\text{O}_3$ (222) and $\alpha\text{-Al}_2\text{O}_3$ (006) is calculated to be 18.21%. However, although the lattice mismatches between both interfaces are relatively high, this does not affect our successful demonstration of single-crystalline group-III-oxide epitaxy structure with the intermediate $\gamma\text{-In}_2\text{O}_3$ layer acting as a wafer-scalable conductive layer, rather than as a buffer layer. Indeed, we envisage that the present work could also lay a foundation for future oxide-based buffer-layer growth, mimicking that of group-III nitrides on sapphire, via other epitaxial growth techniques (e.g., metal-organic CVD, molecular beam epitaxy) that ensure better process controllability and higher growth rate for even better crystal quality.

Figure 3. Cross-sectional high-resolution transmission electron microscopy (HR-TEM) images acquired at (a) $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3$ and (b) $\gamma\text{-In}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ interfaces. Corresponding HR-TEM image, fast Fourier transform (FFT) pattern, and simulated FFT for each individual layer of (c) $\beta\text{-Ga}_2\text{O}_3$, (d) $\gamma\text{-In}_2\text{O}_3$, and (e) $\alpha\text{-Al}_2\text{O}_3$.

Figure 4. (a) Scanning-TEM high-angle annular dark-field (STEM-HAADF) image and (b) corresponding elemental mapping for $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ heterojunction. Simulated crystal structures and corresponding lattice constants for (c) monoclinic $\beta\text{-Ga}_2\text{O}_3$, (d) cubic $\gamma\text{-In}_2\text{O}_3$, and (e) trigonal $\alpha\text{-Al}_2\text{O}_3$.

The stoichiometry and interfaces of each layer can also be determined using RBS, as shown in Figure 5a, where the Ga and In signals overlap and the Ir signal originates simply from the metal layer deposited to prevent electrostatic charging. The simulated elemental concentration profile shown in Figure 5b indicates a thin interdiffusion layer of 30 nm at the $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3$ interface, accounting for approximately 15% of the total thickness in the $\beta\text{-Ga}_2\text{O}_3$ layer, which is unavoidable in high-temperature growth.¹ Based on the RBS model, the stoichiometric ratio for both the $\beta\text{-Ga}_2\text{O}_3$ and $\gamma\text{-In}_2\text{O}_3$ layers is estimated to be 1:1.5.

Figure 5. (a) Measured Rutherford backscattering spectrometry (RBS) spectra for single-crystalline β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterojunction; the inset shows the simulated spectra, which fit with the measured spectra. (b) Percentage of elemental concentration with respect to depth profile as extracted from simulation.

3.3 Energy Band Offset

To evaluate the VBO at the heterojunction of the β -Ga₂O₃/ γ -In₂O₃ interface, the conventional relation due to Kraut et al.⁵¹ was used, which is expressed as:

$$\Delta E_v = (E_{\text{In } 4d}^{\gamma\text{-In}_2\text{O}_3} - E_{\text{VBM}}^{\gamma\text{-In}_2\text{O}_3}) - (E_{\text{Ga } 3d}^{\beta\text{-Ga}_2\text{O}_3} - E_{\text{VBM}}^{\beta\text{-Ga}_2\text{O}_3}) - (E_{\text{In } 4d}^{\gamma\text{-In}_2\text{O}_3} - E_{\text{Ga } 3d}^{\beta\text{-Ga}_2\text{O}_3}) \quad (4)$$

where ΔE_v is the VBO, $E_{\text{In } 4d}^{\gamma\text{-In}_2\text{O}_3}$ is the binding energy at the In 4d core level of the γ -In₂O₃ layer, $E_{\text{VBM}}^{\gamma\text{-In}_2\text{O}_3}$ is the valence-band maximum (VBM) of the γ -In₂O₃ layer, $E_{\text{Ga } 3d}^{\beta\text{-Ga}_2\text{O}_3}$ is the binding

energy at the Ga 3d core level of the β -Ga₂O₃ layer, and $E_{\text{VBM}}^{\beta\text{-Ga}_2\text{O}_3}$ is the VBM of the β -Ga₂O₃ layer. Meanwhile, the CBO (ΔE_c) is evaluated as:

$$\Delta E_c = \left(E_g^{\beta\text{-Ga}_2\text{O}_3} - E_g^{\gamma\text{-In}_2\text{O}_3} \right) - \Delta E_v \quad (5)$$

where $E_g^{\beta\text{-Ga}_2\text{O}_3}$ and $E_g^{\gamma\text{-In}_2\text{O}_3}$ are the optical bandgaps of β -Ga₂O₃ and γ -In₂O₃, respectively.

Figure 6a shows the XPS spectra of the In 4d core level and the VBM of a bulk γ -In₂O₃ sample, corresponding to binding energies of 17.47 and 2.05 ± 0.05 eV, respectively; thus, the difference in binding energy (i.e., $E_{\text{In } 4d}^{\gamma\text{-In}_2\text{O}_3} - E_{\text{VBM}}^{\gamma\text{-In}_2\text{O}_3}$) is calculated as 15.42 eV. Similarly, Figure 6b shows the XPS spectra of the Ga 3d core level and the VBM of a bulk β -Ga₂O₃ sample, with binding energies of 19.90 and 2.96 ± 0.05 eV, respectively, resulting in a difference in binding energy (i.e., $E_{\text{Ga } 3d}^{\beta\text{-Ga}_2\text{O}_3} - E_{\text{VBM}}^{\beta\text{-Ga}_2\text{O}_3}$) of 6.94 eV. The binding energies of the Ga 3d and In 4d core-level spectra, as examined from a thin β -Ga₂O₃ layer grown on a γ -In₂O₃ sample, are shown in Figure 6c; these are 19.90 and 17.00 eV, respectively, and the energy difference (i.e., $E_{\text{In } 4d}^{\gamma\text{-In}_2\text{O}_3} - E_{\text{Ga } 3d}^{\beta\text{-Ga}_2\text{O}_3}$) is thus -2.9 eV. With that, the VBO calculated using Equation (4) is $\Delta E_v = 1.38$ eV.

To establish the necessary parameters for energy-band alignment at the β -Ga₂O₃/ γ -In₂O₃ interface, Figures 6d and 6e show the reflection electron energy loss spectra (REELS) of bulk γ -In₂O₃ and β -Ga₂O₃ samples. The optical bandgaps extracted from REELS were 3.65 eV for the γ -In₂O₃ sample and 4.87 eV for the β -Ga₂O₃ sample. Substituting these optical bandgaps into Equation (5) gives $\Delta E_c = |0.16 \text{ eV}|$. The detailed energy-band diagram of the single-crystalline PLD-grown β -Ga₂O₃/ γ -In₂O₃ interface, as deduced from the above measurements and calculations, is shown in Figure 6f, yielding a type-II heterojunction.

Figure 6. Energy-band alignment at single-crystalline β -Ga₂O₃/ γ -In₂O₃ heterojunction: (a) In 4d core-level and valence-band spectra for bulk γ -In₂O₃ sample; (b) Ga 3d core-level and valence-band spectra for bulk β -Ga₂O₃ sample; (c) Ga 3d and In 4d core levels for β -Ga₂O₃/ γ -In₂O₃ heterojunction sample; reflection electron energy loss spectra of (d) γ -In₂O₃ and (e) β -Ga₂O₃ sample; (f) schematic of energy-band alignment at single-crystalline β -Ga₂O₃/ γ -In₂O₃ heterojunction.

3.4 Photoelectrical Performance

Figure 7a shows the I - V characteristics of the vertically structured single-crystalline β -Ga₂O₃/ γ -In₂O₃ photodetector fabricated on the α -Al₂O₃ platform with an area of 0.25 mm² under dark and illuminated conditions from 230 to 300 nm. At zero bias, the dark current of the photodetector is low at 1.65×10^{-9} A, increasing substantially to 7.26×10^{-6} A at a bias voltage of -5 V. Based on the measured I - V characteristics, the responsivity (R_λ) with respect to the illumination wavelength can be calculated as:

$$R_\lambda = \eta_\lambda \frac{q\lambda}{hc} = \frac{I_\lambda - I_{\text{dark}}}{P_\lambda} \quad (6)$$

where η_λ is the EQE for a given illumination wavelength, q is the elementary charge, λ is the radiation wavelength, h is Planck's constant, c is the speed of light, I_λ is the photogenerated

current at a specific illumination wavelength, I_{dark} is the dark current, and P_{λ} is the illuminated optical power on the photodetector. Based on Equation (6), the EQE is determined as:

$$\eta_{\lambda} = R_{\lambda} \times \frac{hc}{q\lambda} \quad (7)$$

The corresponding responsivity and EQE of the photodetector measured at a bias voltage of -5 V are shown in Figure 7b, where the peak response wavelength is at 250 nm with a responsivity of 94.3 A/W and an EQE of $4.6 \times 10^4\%$. The response spectrum reduces strategically toward the UVB wavelength region (i.e., >280 nm), thereby elucidating the potential for a true solar-blind wavelength-selective photodetector with a measured UVC-to-UVB rejection ratio (R_{250}/R_{280}) of 7.69. The common long absorption tail observed after the bandgap energy of the β -Ga₂O₃ absorption layer (i.e., 4.87 eV) can be attributed to near-surface defects⁵² and the deep levels formed around the Fermi level because of oxygen vacancies,⁵³ thereby affecting the absorption properties of the β -Ga₂O₃ layer. Furthermore, the specific detectivity (D^*) of the photodetector, which typically governs the signal-to-noise ratio,⁵⁴ can be calculated as:

$$D_{\lambda}^* = \frac{(R_{\lambda})(\sqrt{A})}{\sqrt{2qI_{\text{dark}}}} \quad (8)$$

where A [cm²] is the total illuminated area. Figure 7c shows the D^* of the single-crystalline β -Ga₂O₃/ γ -In₂O₃ photodetector in the measured wavelength range of 230–300 nm, with the peak D^* of 3.09×10^{12} Jones at 250 nm under the bias voltage of -5 V. The high external quantum efficiency and multiplication gain exhibited by our photodetector can be correlated with the trapping and de-trapping of photogenerated carriers arising from the deep-trap states formed inherently by the oxygen vacancies typically observed in Ga₂O₃-based photodetectors.^{55–57}

Figure 7. Photoelectrical performance of fabricated deep-UV photodetector based on single-crystalline β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterojunction: (a) I - V characteristics under dark and illuminated conditions with illumination wavelengths of 230–300 nm; calculated (b) responsivity, external quantum efficiency (EQE), and (c) specific detectivity (D^*) spectra for 230–300 nm under bias of -5 V; (d) time response characteristics measured at peak responsivity of 250 nm and bias voltage of -5 V; measured and bi-exponential fitting curves corresponding to (e) rise time and (f) decay time.

The time response characteristics of the β -Ga₂O₃/ γ -In₂O₃ photodetector are shown in Figure 7d for the peak response wavelength of 250 nm, a power density of $124 \pm 6 \mu\text{W}/\text{cm}^2$, and a bias voltage of -5 V. The corresponding measured and fitted curves of the rise (τ_{rise}) and decay (τ_{decay}) times are shown in Figures 7e and 7f. A bi-exponential function as given by Equation (9) was used to estimate both the rise and decay time:

$$I = I_0 + Ae^{-t/\tau_{d1}} + Be^{-t/\tau_{d2}} \quad (9)$$

where I is the measured current, I_0 is the steady-state current, and A and B are the fitting constants corresponding to the fast (τ_{d1}) and slow (τ_{d2}) components. In terms of the rise time, as shown in Figure 7e, based on Eq. (9), it revealed the fast and slow rise time of $\tau_{d1} = 0.596 \pm 0.003$ s and $\tau_{d2} = 3.221 \pm 0.035$ s. The decay time, on the other hand, revealed the fast and slow decay components of $\tau_{d1} = 0.218 \pm 0.002$ s and $\tau_{d2} = 10.868 \pm 0.125$ s, in which the fast component is

attributed to the direct band-to-band recombination process while the slow component is governed by the trapping and de-trapping process due to various defect states present in the Ga_2O_3 absorption layer.^{55,56}

Table 1 summarizes the photoelectrical performances of photodetectors fabricated based on various Ga_2O_3 -based heterojunctions, in which the proposed $\beta\text{-Ga}_2\text{O}_3/\gamma\text{-In}_2\text{O}_3$ photodetector on an $\alpha\text{-Al}_2\text{O}_3$ platform outperforms substantially in terms of responsivity and EQE, mainly because of its single-crystallinity and the inclusion of the high-quality conductive intermediate $\gamma\text{-In}_2\text{O}_3$ layer with low energy-band offset, ideal for effective photocarrier transport. Compared to previous work as presented in Table 1 and Figure 8, the achieved peak responsivity (i.e., 94.3 A/W) and EQE (i.e., $4.6 \times 10^4\%$) in our vertical-type photodetector are higher than those for Ga_2O_3 grown on a conductive layer or substrate (e.g., SiC,⁵⁸ Nb:SrTiO₃,⁵⁵ ZnO,⁵⁹ graphene,³⁵ GaN,⁶⁰ NiO¹) forming the heterojunction. The achieved performance is also closer to that of a similar vertical-type structure based on a highly conductive $p\text{-Si}$ substrate,⁶¹ which however does not exhibit the required transparency across the UV-to-visible wavelength region for a transparent-oxide-based platform. Moreover, in terms of response time, our achieved rise time (i.e., 0.596 s) and decay time (i.e., 0.218 s) in τ_{d1} are faster than those of other $\beta\text{-Ga}_2\text{O}_3$ grown on a thick conductive layer or substrate (e.g., $p\text{-Si}$,⁶¹ Nb:SrTiO₃,⁵⁵ ZnO⁵⁹), thereby increasing the carrier transition time significantly. In the present work, with the inclusion of the conductive intermediate $\gamma\text{-In}_2\text{O}_3$ layer and the design of a vertical-type photodetector structure, the response times were reduced because of the rather thin $\beta\text{-Ga}_2\text{O}_3$ -based absorption layer (i.e., ~350 nm), allowing the photogenerated carriers to be transported effectively to the metal pads without significant delays. However, note that the slow components (τ_{d2}) of the rise and decay times may still require significant improvements because of various defect states arising from the material quality of the absorption layer and can be improved in the future with an optimized growth process. It is also important to note that our work surpasses other previous work, particularly in terms of the response time, demonstrated for metal–semiconductor–metal-based $\beta\text{-Ga}_2\text{O}_3$ photodetectors grown directly on an Al_2O_3 substrate with no intermediate conductive layer, which typically suffer from long decay times in excess of 0.3 s,^{62,63} because of the limitation regarding the distance between the interdigitated electrodes, which increases the carrier transition time, as well as the use of a non-conductive Al_2O_3 substrate that limits the feasibility for a vertical-type photodetector structure.

Table 1. Performance comparison across previously demonstrated Ga₂O₃-based heterojunction photodetectors.

Type of Heterojunction	Peak Responsivity (A/W)	EQE (%)	Specific Detectivity (Jones)	Response Time (s)		Ref.
				Rise (τ_{d1}/τ_{d2})	Decay (τ_{d1}/τ_{d2})	
<i>β-Ga₂O₃/SiC</i>	0.068 (225 nm)	38	-	0.0012	0.0015	58
<i>β-Ga₂O₃/p-Si</i>	370 (254 nm)	1.8×10^5	-	1.79/4.54	0.27/2.72	61
<i>β-Ga₂O₃/Nb:SrTiO₃</i>	43.31 (254 nm)	2.1×10^4	-	1.08/8.78	0.65/6.87	55
<i>β-Ga₂O₃/ZnO</i>	0.35 (254 nm)	1.7×10^2	-	1.02/11.66	0.62/7.84	59
<i>β-Ga₂O₃/Graphene</i>	12.8 (254 nm)	6.7×10^3	1.3×10^{13}	0.0015*	0.002*	35
<i>Sn:Ga₂O₃/GaN</i>	3.05 (254 nm)	1.5×10^3	1.69×10^{13}	-	0.018/0.148	60
<i>β-Ga₂O₃/p-CuSCN (microwire)</i>	0.0133 (254 nm)	6.49	9.43×10^{11}	0.19/3.63	0.16/0.17	64
<i>β-Ga₂O₃/NiO</i>	0.415 (260 nm)	197.92	2.27×10^{11}	-	-	1
<i>β-Ga₂O₃/γ-In₂O₃</i>	94.3 (250 nm)	4.6×10^4	3.09×10^{12}	0.596/3.221	0.218/10.868	This work

*Measured based on 10% to 90% of the normalized photoresponse curve

Figure 8. Comparison of performance in terms of responsivity and response time for β -Ga₂O₃-based heterojunction photodetector. The response time was taken from the fast decay time (τ_{d1}) in Table 1.

4. CONCLUSIONS

In this work, single-crystalline all-oxide-based β -Ga₂O₃/ γ -In₂O₃/ α -Al₂O₃ heterostructures were presented and investigated fundamentally as vertically structured optoelectronic-device platforms. The orientation relationships related to the single-crystalline heterostructure, ascertained from XRD and HR-TEM analysis, were found to be β -Ga₂O₃ ($\bar{2}01$) $\parallel$ γ -In₂O₃ (222) $\parallel$ α -Al₂O₃ (003) and β -Ga₂O₃ (020) $\parallel$ γ -In₂O₃ ($\bar{2}02$) $\parallel$ α -Al₂O₃ (300). The energy-band offsets related to the β -Ga₂O₃/ γ -In₂O₃ interface reveal a type-II heterojunction with CBO and VBO values of 0.16 and 1.38 eV, respectively. Including a wafer-scalable intermediate TCO layer of single-crystalline γ -In₂O₃ therefore ensured high carrier transport, visible transparency, and antioxidation properties for future oxide-based platforms. The vertically structured photodetector realized based on the α - γ - β heterostructure also exhibited high responsivity (94.3 A/W) and external quantum efficiency (4.6 $\times$ 10⁴%), ideal for sensitive UVC photodetection applications such as flame and oil-leak detection.

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Notes

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