The Effect of a Nucleation Layer on Morphology and Grain Size in MOCVD-Grown $\beta$-$\text{Ga}_2\text{O}_3$ Thin Films on C-Plane Sapphire

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Abstract: $\beta$-$\text{Ga}_2\text{O}_3$ thin films grown on widely available c-plane sapphire substrates typically exhibit structural defects due to significant lattice and thermal expansion mismatch, which hinder the use of such films in electronic devices. In this work, we studied the impact of a nucleation layer on MOCVD-grown $\beta$-$\text{Ga}_2\text{O}_3$ thin film structure and morphology on a c-plane sapphire substrate. The structure and morphology of the films were investigated by X-ray diffraction, atomic force microscopy, transmission and scanning electron microscopy, while the composition was confirmed by X-ray photoelectron spectroscopy and micro-Raman spectroscopy. It was observed that the use of a nucleation layer significantly increases the grain size in the films in comparison to the films without, particularly in the samples in which $\text{H}_2\text{O}$ was used alongside $\text{O}_2$ as the oxygen source for the nucleation layer growth. Our study demonstrates that a nucleation layer can play a critical role in obtaining high quality $\beta$-$\text{Ga}_2\text{O}_3$ thin films on c-plane sapphire.

Keywords: gallium oxide; MOCVD; thin films; UWBG materials; nucleation layer

1. Introduction

Gallium oxide ($\text{Ga}_2\text{O}_3$) has recently attracted significant scientific attention as a new, promising ultra-wide bandgap semiconductor [1,2]. Several $\text{Ga}_2\text{O}_3$ polymorphs exist of which monoclinic $\beta$-$\text{Ga}_2\text{O}_3$ is the thermodynamically most stable phase [3]. It has a 4.6–4.9 eV wide bandgap [4]; therefore, the theoretical limit value of its breakdown voltage exceeds those of the more commonly studied SiC and GaN [5]. Together with its high mechanical strength and chemical and thermal stability [4], $\beta$-$\text{Ga}_2\text{O}_3$ properties show promise for a variety of applications in power electronics [5–7] and solar-blind deep-ultraviolet photodetectors [8,9].

$\beta$-$\text{Ga}_2\text{O}_3$ thin films have been grown by a variety of methods, such as pulsed laser deposition [10,11] and magnetron sputtering [12]; however, molecular beam epitaxy [13,14] and metalorganic chemical vapour deposition (MOCVD) [15–18] are the most promising methods for high deposition rates and high-quality device fabrication. MOCVD in particular has the advantage of high growth rates (up to 10 $\mu$m/h) with sub-nanometre surface roughness [16]. Typically, triethylgallium (TEGa) is used as the Ga precursor and $\text{O}_2$ as the oxygen precursor in $\text{Ga}_2\text{O}_3$ MOCVD growth; however, films with a comparable quality have also been grown with the significantly cheaper trimethylgallium (TMGa) [19,20]. Regarding the oxygen precursor, commonly used $\text{O}_2$ is the established choice, yet the use of $\text{N}_2\text{O}$ can lead to a lower background charge carrier concentration [21] and the use of water ($\text{H}_2\text{O}$) vapour can potentially have a positive effect on the kinetic conditions during film growth due to the presence of hydrogen [20]. There are only a few papers reporting the use of $\text{H}_2\text{O}$ as an oxygen precursor; Alema et al. demonstrated a reduction in carrier concentration by adding a small amount of $\text{H}_2\text{O}$ to the $\text{O}_2$ flow during the film growth [22],...
while Lee et al. and Chen et al. reported Ga$_2$O$_3$ film growth using H$_2$O as the oxygen precursor without further discussions on the choice [23–25].

The use of native Ga$_2$O$_3$ substrates leads to the highest quality epitaxial films [10,21]; however, they are still expensive and thus currently uneconomical for large-scale production. On the other hand, α-Al$_2$O$_3$ (sapphire) substrates are widely available, but the significant 6.6% lattice and thermal expansion mismatch between the (0001) c-plane sapphire and β-Ga$_2$O$_3$ [26] leads to structural defects during the growth process such as threading dislocations, which hinder the use of such films in electronic devices. Large numbers of threading dislocations lead to formation of low-angle grain boundaries represented as a configuration of edge dislocations, whose axes lie in the plane of the boundary between the grains. In the case of GaN on c-plane sapphire, the high density of dislocations due to the large (~14%) lattice mismatch was overcome by introducing a buffer layer through controlled nucleation of highly crystalline islands [27]. As for Ga$_2$O$_3$, to the best of our knowledge, there are no dedicated reports on the role of a nucleation or a buffer layer in MOCVD growth. Alema et al. reported deposition of a 30 nm thick low-temperature (650 °C) buffer layer on c-plane sapphire before the film overgrowth in order to improve its crystalline quality [16], and Chen et al. used a 15 nm thick nucleation layer, deposited at 600 °C, to grow ε-Ga$_2$O$_3$ film on c-plane sapphire [28]. However, no comparison of the films grown with and without a nucleation layer has been made. A positive effect of a seed layer was recently demonstrated by Gu et al. [29] for sputter-deposited Ga$_2$O$_3$ films on variously oriented silicon substrates, as well as of α-Cr$_2$O$_3$ buffers for α-Ga$_2$O$_3$ growth by Polyakov et al. [30].

In this work, we investigated and compared MOCVD-grown β-Ga$_2$O$_3$ thin films (A) without any nucleation layer, (B) with a nucleation layer grown by using an O$_2$ precursor as the oxygen source and (C) with a nucleation layer grown by using a combination of H$_2$O and O$_2$ precursors. The focus of this study was the film structure and morphology. It was observed that the use of a nucleation layer significantly increases the grain size in our films, particularly in the samples in which H$_2$O was used alongside O$_2$ in the nucleation layer growth. Further development of a more complex nucleation or buffer layer could lead to significantly higher crystalline quality β-Ga$_2$O$_3$ films on widely available c-plane sapphire wafers.

2. Materials and Methods

Heteroepitaxial β-Ga$_2$O$_3$ thin films were grown on 2-inch c-plane sapphire wafers (Biotain Crystal Co. Ltd., Xiamen, China) using the AIX-200RF (AIXTRON, Herzogenrath, Germany) MOCVD system. Trimethyl gallium (TMGa) was used as the gallium source, while water (H$_2$O) and oxygen (O$_2$) were used as oxygen sources. Nitrogen (N$_2$) was used as a carrier gas with the total flow of 3000 standard cubic centimetres per minute (sccm) during the deposition process. Before growth, the sapphire substrates were annealed for 2 min at 1070 °C in a N$_2$ atmosphere at 200 mbar pressure. The main Ga$_2$O$_3$ layer for all samples was deposited in a N$_2$ atmosphere and 30 mbar pressure at 820 °C temperature. TMGa and O$_2$ flow rates were 120 µmol/min and 20 mmol/min, respectively. For a 30 min growth process the thickness of the as-grown thin films was measured to be around 1.5 µm.

Three types of growth procedures were implemented and studied:

- Sample A was prepared without any nucleation layer.
- Sample B was prepared with a low-temperature (LT) nucleation layer deposited using O$_2$ precursor as the oxygen source. The amorphous LT layer (around 30 nm thick) was deposited at 720 °C for 150 s with a 90 µmol/min TMGa flow rate and a 16 mmol/min O$_2$ flow rate (Step 1 in Figure 1). Then, the sample temperature was raised to 1030 °C to perform high-temperature annealing of the layer (Step 2 in Figure 1), supposedly crystallizing it as suggested by X-ray diffraction measurements in Figure S1 in supplementary materials. After a 150-s-long dwell time, the temperature of the sample was decreased to the main layer growth temperature (see above) followed by the 30 min deposition (Step 3 in Figure 1).
Sample C was prepared with a nucleation layer deposited using both precursors H$_2$O and O$_2$ as the oxygen sources. One LT layer was grown as it was described for Sample B. An additional LT layer was overgrown on top of the first one for 60 s using TMGa and H$_2$O as the gallium and oxygen sources, respectively. The flow rates were 90 µmol/min for TMGa and 320 µmol/min for H$_2$O precursors. Before the growth of the main layer, the temperature was increased to anneal the LT layers similarly as for Sample B.

Figure 1. A schematic of Ga$_2$O$_3$ thin film MOCVD growth steps using a nucleation layer. After the high-temperature annealing of the substrate, a few-nm thick low-temperature Ga$_2$O$_3$ layer is deposited at 720 °C (Step 1). It is then annealed for 150 s at 1030 °C (Step 2), followed by the overgrowth of Ga$_2$O$_3$ film at 820 °C (Step 3).

The thin film surface morphology was examined with a scanning electron microscope (SEM, Thermo Scientific Helios 5 UX, Waltham, MA, USA) and an atomic force microscope (AFM, Veeco, Dimension Edge, Plainview, NY, USA) in tapping mode using a Pointprobe non-contact AFM tip NCHR-50, while the film thickness was determined by ellipsometry (Woollam RC2-XI, Lincoln, NE, USA). X-ray diffraction (XRD, PANalytical, X’Pert Pro, Malvern, UK) patterns were measured using Cu K$_\alpha$ irradiation in Bragg–Brentano Theta–Theta geometry, with a step size of 0.026 deg, tube powered by 40 kV and 30 mA and rocking curves (ω-scans) were measured using parallel beam optics and monochromatic (Cu K$_\alpha$) radiation, with a step size of 0.001 deg. The lamellae were prepared by focused ion beam (FIB, Thermo Scientific Helios 5 UX, Waltham, MA, USA) to investigate the cross-section of the thin film with a transmission electron microscope (TEM, FEI Tecnai G20, Hillsboro, OR, USA) operated at 200 kV. The thin film was coated with a 30 nm thick Au layer and a 1.5 µm thick Pt layer to protect the surface from Ga$^+$ exposure during FIB preparation. Micro-Raman spectroscopy measurements were performed using a TriVista 777 confocal Raman system (Princeton Instruments, Trenton, NJ, USA, 750 mm focal length, 1800 lines/mm grating) equipped with an Olympus microscope and Ulls 2 MPlanN 100 × /0.90 objective, a continuous-wave diode-pumped laser Cobolt Samba 150 (HÜBNER Photonics, Kassel, Germany) (λ = 532 nm) and Andor iDus DV420A-OE CCD camera (Andor Technology, Belfast, Northern Ireland). The X-ray photoelectron spectrometry (XPS) measurements were performed by an ESCALAB 250Xi (ThermoFisher, Waltham, MA, USA) instrument to determine the film chemical composition. An Al K$_\alpha$ X-ray tube with the energy of 1486 eV was used as an excitation source, the size of the analysed sample area was 650 µm × 100 µm and the angle between the analyser and the sample surface was 90°. An electron gun was used to perform charge compensation. The base pressure during the spectra acquisition was better than 10$^{-7}$ mbar.

3. Results and Discussion

The surface morphology of the as-deposited Ga$_2$O$_3$ thin films was investigated with AFM and SEM and is shown in Figure 2. It can be observed that the size of the crystalline
domains or grains significantly increases when a nucleation layer is used. The sample without any nucleation layer (Sample A) exhibits a small-grained (sub-micron) structure without any visually distinguishable orientation, while the use of a nucleation layer in Sample B and Sample C seems to lead to much larger grains. Furthermore, the use of a H₂O precursor alongside O₂ in the nucleation layer growth in Sample C increases the grain size in the overgrown Ga₂O₃ film even further. During the establishment of the film growth process, a series of films using various O₂ and H₂O precursor ratios were deposited and it was observed that increasing the proportion of O₂ in the precursor flow leads to a higher growth rate and larger grains (see SEM images in Figure S2 in supplementary materials); however, it also favours growth of grains of crystalline orientations other than (−201), as can be seen in the XRD patterns in Figure S3 in supplementary materials. This is caused by the intensive parasitic reaction between TMGa and O₂ precursors. In order to reduce the influence of the parasitic reaction, a low process pressure (below 50 mBar) was used. By increasing the pressure above 50 mBar, the film deposition rate decreases drastically. However, in the case of the H₂O precursor, even 200 mBar pressure during growth does not affect the deposition rate. Therefore, the use of H₂O during the nucleation layer growth in Sample C seems to reduce the misorientation of grains in the initial growth step, which consequently leads to fewer structural defects in the overgrown film and thus, larger grains. The mean grain size was calculated from processing several AFM measurements for each sample. Regarding the surface roughness, maximum peak height values, Rp, were determined from the AFM measurements and calculated to be 178.2 nm (Sample A), 202.9 nm (Sample B) and 367.6 nm (Sample C). The increase in the surface roughness can be attributed to the formation of larger single-crystalline grains, which tend to protrude.

The crystalline structure of the prepared films was studied with XRD and TEM. A typical XRD pattern (2θ scan) of the films can be seen in Figure 3a, showing a highly crystalline monoclinic β-Ga₂O₃ phase (ICDD-PDF #41-1103) oriented along [−201] direction. Since all the samples gave qualitatively similar patterns, only one is shown here. Rocking curves (ω-scans) of the (−201) peak for all prepared samples showed similar full-width half-maximum (FWHM) values in the range of 1.82–1.93°, which is comparable to the best

![Figure 2. Atomic force microscopy (10 µm × 10 µm scans) and scanning electron microscopy images of (a,b) Sample A, (c,d) Sample B and (e,f) Sample C, respectively, showing the surface morphology of the as-grown Ga₂O₃ films.](image-url)
reported values in the literature for $\beta$-Ga$_2$O$_3$ growth on c-plane sapphire [31,32]. Furthermore, lamellas were prepared in SEM-FIB in order to perform the film cross-section studies with TEM and reveal the crystalline structure of the films. The Ga$_2$O$_3$/sapphire interface can be seen in Figure 3b, confirming the oriented growth of the film. Interplanar distance ($d$-spacing) of the resolved atomic planes in the Ga$_2$O$_3$ film was measured to be 4.73 Å, which matches well with the $[−201]$ growth direction [33]. The analysis of the selected area electron diffraction (SAED) pattern in Figure 3c also supports this. SEM images of the cross-sections (see Figure 3d for Sample C and Figure S4 in supplementary materials for Samples A and B) show columnar structures with different contrast, which could be visible due to slightly misoriented grains. Grain boundaries visible in the cross-section cuts (see Figure 3e) are typically low-angle grain boundaries with misalignment of 1–3°; however, regions of grain misalignment of up to 18° could be occasionally identified in Sample A and Sample B (see Figure S4). The characteristic quantities for the film structure and morphology are summarised in Table 1.

**Table 1.** Characteristic parameters for the as-grown $\beta$-Ga$_2$O$_3$ film structure and morphology.

| Sample | Maximum peak height (roughness) Rp, nm | Mean grain size, μm | Rocking curve FWHM, degrees |
|--------|---------------------------------------|---------------------|----------------------------|
| A      | 178.2                                 | 0.389               | 1.93                       |
| B      | 202.9                                 | 1.502               | 1.87                       |
| C      | 367.6                                 | 1.989               | 1.82                       |

**Figure 3.** (a) A typical X-ray diffraction pattern of a Ga$_2$O$_3$ film on c-plane sapphire, all samples gave qualitatively similar patterns. The inset shows rocking curves ($\omega$-scans) of the $[−201]$ peak of the studied samples. (b) A transmission electron microscope image of the Ga$_2$O$_3$/sapphire interface in Sample C, showing the Ga$_2$O$_3$ growth direction and interplanar spacing, with (c) selected area electron diffraction pattern of the Ga$_2$O$_3$ film. (d) Scanning electron image of the cross-section cut of Sample C. (e) An example of a low-angle grain boundary typically observed in Sample C.
The film composition and purity were verified with micro-Raman spectroscopy and XPS. The Raman spectrum of a Ga$_2$O$_3$ film is shown in Figure 4a together with a pure sapphire substrate spectrum for comparison. The sapphire substrate peaks are marked with ‘S’, while the rest can be attributed to multiple phonon modes with $A_g$ and $B_g$ symmetry of β-Ga$_2$O$_3$, closely matching the spectral positions reported previously in the literature [34]. The XPS survey scan (see Figure 4b) only indicated the presence of Ga and O elements in stoichiometry closely matching the Ga$_2$O$_3$ compound, as well as surface carbon contaminants. The spectra were calibrated to the adventitious C1s peak at 285.0 eV binding energy. No other elements were detected. The high-resolution spectra of Ga3d and O1s peaks are shown in the inset. The Ga3d peak was located at 20.0 eV with an overlapping O2s peak at around 23.5 eV, while the O1s peak was measured to be at 530.8 eV with a shoulder contribution from the C-O chemical state in the organic surface contaminations. The Ga and O peak binding energy values correspond to the Ga$_2$O$_3$ compound as expected [35].

| Sample  | Sample B | Sample C |
|---------|----------|----------|
| Maximum peak height (roughness) $R_p$, nm | 178.2 | 202.9 | 367.6 |
| Mean grain size, µm | 0.389 | 1.502 | 1.989 |
| Rocking curve FWHM, degrees | 1.93 | 1.87 | 1.82 |

**Table 1.** Characteristic parameters for the as-grown β-Ga$_2$O$_3$ film structure and morphology.

**4. Conclusions**

In this study, we demonstrated an improvement in grain size and morphology of MOCVD-grown β-Ga$_2$O$_3$ thin films on c-plane sapphire by using a nucleation layer in the initial film growth step. The structure and morphology of the films were investigated by XRD, TEM, SEM and AFM, and the composition was confirmed by XPS and micro-Raman spectroscopy. The films in which a H$_2$O precursor was used alongside O$_2$ as the oxygen source in the nucleation layer growth exhibit larger grains and slightly better crystallinity in comparison to the films with an O$_2$ only nucleation layer. This is possibly due to less intensive parasitic reactions between TMGa and H$_2$O in comparison to TMGa and O$_2$ precursors, causing fewer structural defects in the nucleation layer and thus larger and highly oriented crystalline grains in the overgrown β-Ga$_2$O$_3$ film. Our study demonstrates
that a nucleation layer can play a critical role in obtaining high quality $\beta$-Ga$_2$O$_3$ thin films on c-plane sapphire, therefore further development of a more complex nucleation or buffer layer recipe could lead to significantly higher crystalline quality $\beta$-Ga$_2$O$_3$ films on widely available c-plane sapphire substrates.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/ma15238362/s1, Figure S1: X-ray diffraction patterns of the nucleation layers after annealing at 1030 $^\circ$C for 150 s. The layers were prepared with (a) O$_2$ and (b) O$_2$ + H$_2$O as the oxygen precursors. The low-temperature layers deposited at 720 $^\circ$C before the annealing are X-ray amorphous. The unmarked peaks belong to the c-plane sapphire substrate., Figure S2: Scanning electron microscope images of MOCVD-grown Ga$_2$O$_3$ thin films using H$_2$O vapour and O$_2$ as oxygen precursors at various ratios. Total oxygen precursor flow was kept constant at 500 sccm. Higher O$_2$ proportion leads to larger grain sizes. Scale bars correspond to 2 $\mu$m., Figure S3: X-ray diffraction patterns of MOCVD-grown Ga$_2$O$_3$ thin films using H$_2$O vapour and O$_2$ as oxygen precursors at various ratios. Total oxygen precursor flow was kept constant at 500 sccm., Figure S4: Scanning electron images of cross-section cuts of (a) Sample A and (c) Sample B. Transmission electron microscope images of the studied grain boundaries in (b) Sample A and (d) Sample B. Typically, most boundaries found in the samples are low-angle grain boundaries with misalignment of 1–3°, however, regions of grain misalignment up to $18^\circ$ could be occasionally identified.

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**References**

1. Shi, J.; Zhang, J.; Yang, L.; Qu, M.; Qi, D.; Zhang, K.H.L. Wide Bandgap Oxide Semiconductors: From Materials Physics to Optoelectronic Devices. *Adv. Mater.* **2021**, 33, e2006230. [CrossRef] [PubMed]
2. Chi, Z.; Asher, J.J.; Jennings, M.R.; Chikoidze, E.; Pérez-Tomás, A. Ga$_2$O$_3$ and Related Ultra-Wide Bandgap Power Semiconductor Oxides: New Energy Electronics Solutions for CO$_2$ Emission Mitigation. *Materials* **2022**, 15, 1164. [CrossRef] [PubMed]
3. Bosi, M.; Mazzolini, P.; Seravalli, L.; Fornari, R. Ga$_2$O$_3$ polymorphs: Tailoring the epitaxial growth conditions. *J. Mater. Chem. C* **2020**, 8, 10975–10992. [CrossRef]
4. Pearton, S.J.; Yang, J.; Cary, P.H., IV; Ren, F.; Kim, J.; Tadjer, M.J.; Mastro, M.A. A review of Ga$_2$O$_3$ materials, processing, and devices. *Appl. Phys. Rev.* **2018**, 5, 011301. [CrossRef]
5. Green, A.J.; Speck, J.; Xing, G.; Moens, P.; Allerstam, F.; Gumaelius, K.; Higashiwaki, M. $\beta$-Gallium oxide power electronics. *Appl. Mater.* **2022**, 10, 029201. [CrossRef]
6. Pearton, S.J.; Ren, F.; Tadjer, M.; Kim, J. Perspective: Ga$_2$O$_3$ for ultra-high power rectifiers and MOSFETS. *J. Appl. Phys.* **2018**, 124, 220901. [CrossRef]
7. Zhang, J.; Dong, P.; Dang, K.; Zhang, Y.; Yan, Q.; Xiang, H.; Su, J.; Liu, Z.; Si, M.; Gao, J.; et al. Ultra-wide bandgap semiconductor Ga$_2$O$_3$ power diodes. *Nat. Commun.* **2022**, 13, 3900. [CrossRef] [PubMed]
8. Varshney, U.; Aggarwal, N.; Gupta, G. Current advances in solar-blind photodetection technology: Using Ga2O3 and AlGaN. *J. Mater. Chem. C* **2022**, *10*, 1573–1593. [CrossRef]
9. Kaur, D.; Kumar, M. A Strategic Review on Gallium Oxide Based Deep-Ultraviolet Photodetectors: Recent Progress and Future Prospects. *Adv. Opt. Mater.* **2021**, *9*, 2002160. [CrossRef]
10. Jeon, H.M.; Leedy, K.D.; Look, D.C.; Chang, C.S.; Muller, D.A.; Badescu, S.C.; Chabak, K.D. Homoepitaxial β-Ga2O3 transparent conducting oxide with conductivity α = 2323 S cm⁻¹. *Appl. Mater. 2021*, *9*, 101105. [CrossRef]
11. Khartsev, S.; Nordell, N.; Hammar, M.; Purans, J.; Hallen, A. High-Quality Si-Doped β-Ga2O3 Films on Sapphire Fabricated by Pulsed Laser Deposition. *Phys. Status Solidi Basic Res.* **2021**, *258*, 2–6.
12. Siikumar, A.K.; Nehate, S.D.; Sundaram, K.B. Review: RF Sputtered Films of Ga2O3 Thin Films for Supercapacitors Applications. *J. Appl. Phys.* **2020**, *109*, 031102. [CrossRef]
13. Vogt, P.; Hensling, F.; Azizie, K.; Chang, C.; Turner, D.; Park, J.; McCandless, J. Adsorption-controlled growth of Ga2O3 by suboxide molecular-beam epitaxy. *Appl. Mater. 2017*, *10*, 051102. [CrossRef]
14. Krishnamoorthy, S.; Xia, Z.; Bajaj, S.; Brenner, M.; Rajan, S. Delta-doped β-gallium oxide field-effect transistor. *Appl. Phys. Express* **2019**, *12*, 012504. [CrossRef]
15. Zhang, Y.; Alema, F.; Mauze, A.; Koksal, O.S.; Miller, R.; Osinsky, A.; Speck, J.S. MOCVD grown epitaxial β-Ga2O3 thin film with an electron mobility of 176 cm²/Vs at room temperature. *Appl. Mater. 2019*, *7*, 022506. [CrossRef]
16. Alema, F.; Hertog, B.; Osinsky, A.; Mukhopadhyay, P.; Toporkov, M.; Schoenfeld, W.V. Fast growth rate of epitaxial β-Ga2O3 by close coupled showerhead MOCVD. *J. Cryst. Growth 2017*, *475*, 77–82. [CrossRef]
17. Kjapsna, A.; Dimitrocenko, L.; Tale, I.; Trukhin, A.; Ignatans, R.; Grants, R. Characterization of crystalline structure and morphological properties of Ga2O3 thin film grown by MOCVD technique. *Key Eng. Mater.* **2017**, *721*, 253–257. [CrossRef]
18. Feng, Z.; Anhar Uddin Bhuiyan, A.F.M.; Karim, M.R.; Zhao, H. MOCVD homoepitaxy of Si-doped (010) β-Ga2O3 thin films with superior transport properties. *Appl. Phys. Lett.* **2019**, *114*, 250601. [CrossRef]
19. Seryogin, G.; Seryogin, G.; Alema, F.; Valente, N.; Fu, H.; Steinbrunner, E.; Neal, A.T.; Osinsky, A. MOCVD growth of high purity Ga2O3 epitaxial films using trimethylgallium precursor. *Appl. Phys. Lett.* **2020**, *117*, 262101. [CrossRef]
20. Wagner, G.; Baldini, M.; Gogova, D.; Schmidbauer, M.; Schewski, R.; Albrecht, M.; Fornari, R. Homoepitaxial growth of β-Ga2O3 layers by metal-organic vapor phase epitaxy. *Phys. Status Solidi Appl. Mater. Sci.* **2014**, *211*, 27–33. [CrossRef]
21. Alema, F.; Alema, F.; Zhang, Y.; Osinsky, A.; Orishchin, N.; Valente, N.; Mauze, A.; Speck, J.S. Low 10–14 cm⁻³ free carrier concentration in epitaxial β-Ga2O3 grown by MOCVD. *Appl. Mater. 2020*, *8*, 021110. [CrossRef]
22. Alema, F.; Zhang, Y.; Mauze, A.; Itoh, T.; Speck, J.S.; Hertog, B.; Osinsky, A. H2O vapor assisted growth of β-Ga2O3 by MOCVD. *AIP Adv.* **2020**, *10*, 085002. [CrossRef]
23. Lee, J.; Kim, H.; Gautam, L.; He, K.; Hu, X.; Dravid, V.; Razeghi, M. Study of Phase Transition in MOCVD Grown Ga2O3 from α to β Phase by Ex Situ and In Situ Annealing. *Photonics 2021*, *8*, 17. [CrossRef]
24. Lee, J.; Kim, H.; Gautam, L.; Razeghi, M. Highly Conductive Co-Doped Ga2O3:Si-In Grown by MOCVD. *Coatings 2021*, *11*, 287. [CrossRef]
25. Chen, W.; Chen, Z.; Li, Z.; Fei, Z.; Pei, Y.; Wang, G.; He, Z. Heteroepitaxy of ε-Ga2O3 thin films grown on AlN/Si(111) templates by metal-organic chemical vapor deposition. *Appl. Surf. Sci.* **2022**, *581*, 152335. [CrossRef]
26. Nakagomi, S.; Kokubun, Y. Crystal orientation of β-Ga2O3 thin films formed on c-plane and a-plane sapphire substrate. *J. Cryst. Growth 2012*, *349*, 12–18. [CrossRef]
27. Gibart, P. Metal organic vapour phase epitaxy of GaN and lateral overgrowth. *Reports Prog. Phys.* **2004**, *67*, 667. [CrossRef]
28. Chen, Z.; Li, Z.; Zhuo, Y.; Chen, W.; Ma, X.; Pei, Y.; Wang, G. Layer-by-layer growth of ε-Ga2O3 thin film by metal–organic chemical vapor deposition. *Appl. Phys. Express 2018*, *11*, 101101. [CrossRef]
29. Gu, K.; Zhang, Z.; Tang, K.; Huang, J.; Shang, Y.; Shen, Y.; Liao, M.; Wang, L. Effect of a seed layer on microstructure and electrical properties of Ga2O3 films using variously oriented Si substrates. *Vacuum 2020*, *222*, 110671. [CrossRef]
30. Polyakov, A.; Nikolaev, V.; Stepanov, S.; Almaev, A.; Pechnikov, A.; Yakimov, E.B.; Kushnarev, B.O.; Schemerov, I.V.; Scheglov, M.; Chernykh, A.V.; et al. Effects of sapphire substrate orientation on Sn-doped α-Ga2O3 grown by halide vapor phase epitaxy using α-Ga2O3 buffers. *J. Phys. D. Appl. Phys. 2022*, *55*, 495102. [CrossRef]
31. Gogova, D.; Ghezellou, M.; Tran, D.Q.; Richter, S.; Papamichail, A.; Hassan, J.U.; Darakchieva, V. Epitaxial growth of β-Ga2O3 by hot-wall MOCVD. *AIP Adv.* **2022**, *12*, 055022. [CrossRef]
32. Rafique, S.; Han, L.; Neal, A.T.; Mou, S.; Tadjer, M.J.; French, R.H.; Zhao, H. Heteroepitaxy of N-type β-Ga2O3 thin films on sapphire substrate by low pressure chemical vapor deposition. *Appl. Phys. Lett.* **2016**, *109*, 132103. [CrossRef]
33. Åhman, J.; Svensson, G.; Albertsson, J. A Reinvestigation of β-Gallium Oxide. *Acta Crystallogr. Sect. C Cryst. Struct. Commun.* **1996**, *52*, 1336–1338. [CrossRef]
34. Kranert, C.; Sturm, C.; Schmidt-Grund, R.; Grundmann, M. Raman tensor elements of β-Ga2O3. *Sci. Rep.* **2016**, *6*, 35964. [CrossRef] [PubMed]
35. Ghose, S.; Rahman, S.; Hong, L.; Rojas-Ramirez, J.S.; Jin, H.; Park, K.; Droopad, R. Growth and characterization of β-Ga2O3 thin films by molecular beam epitaxy for deep-UV photodetectors. *J. Appl. Phys.* **2017**, *122*, 095302. [CrossRef]