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Effect of Substrate Temperature on Structure, Morphology and Optical Properties of β-Ga_2O_3 Thin Film Grown on GaN by MBE

Boyuan Feng[1,3], Gaohang He[1]*, Zhengcheng Li[1,3], Ying Wu[1], Zhongming Zeng[2] and Sunan Ding[1,3]

[1]Vacuum Interconnected Nanotech Workstation (Nano-X), Suzhou Institute of Nano-Tech and Nano-Bionics (SINANO), China

[2]Key Laboratory of Nanodevices and Applications, Suzhou Institute of Nano-Tech and Nano-Bionics, China

[3]School of Nano-Tech and Nano-Bionics, University of Science and Technology of China, China

Research Article

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***Corresponding author:** Gaohang He, Vacuum Interconnected Nanotech Workstation (Nano-X), Suzhou Institute of Nano-Tech and Nano-Bionics (SINANO), Chinese Academy of Sciences (CAS), Suzhou 215123, China, Email: ghhe2018@sinano.ac.cn

Abstract

The structure, morphology and optical properties of β-Ga_2O_3 thin films grown on GaN at various substrate temperature by ozone molecular beam epitaxy (MBE) are investigated in this work. (-201)-oriented β-Ga_2O_3 thin films are formed on c-plane GaN template substrate. When the substrate temperature increases, the crystal quality of β-Ga_2O_3 thin film improves, and a high-crystalline-quality β-Ga_2O_3 thin film is obtained at the substrate temperature of 700 °C. The Φ scans of X-ray diffraction is utilized to characterize the β-Ga_2O_3 thin film, from the result we find that the β-Ga_2O_3 thin film has a six-fold domain structure, attributed to the epitaxial relationships (β-Ga_2O_3 [010] // GaN [11-20] and β-Ga_2O_3 [102] // GaN [1-100]). Base on the morphology, it can be seen that the β-Ga2O3 thin film follows the island-growth model, and the size of the island increases as the substrate temperature increases. Furthermore, it is found that the defect related luminescence decreases with the increase of substrate temperature by analyzing the CL spectra, implying the improvement of crystal quality. The presented optimized β-Ga2O3 thin film grown on GaN template substrate should effectively promote the development of high reliable performance self-powered ultraviolet (UV) photodetector based on the Ga2O3/GaN heterojunction.

Keywords: β-Ga_2O_3; MBE; GaN Template Substrate; Temperature Influence

Abbreviations: UV: Ultraviolet; SBDs: Schottky Barrier Diodes; MESFETs: Metal-Semiconductor Field-Effect-Transistors; MOSFETs: Metal-Oxide-Semiconductor Field-Effect-Transistors; PLD: Pulse Laser Deposition; MBE: Molecular Beam Epitaxy; MOCVD: Metal Organic Chemical Vapor Deposition; XRD: X-Ray Diffraction; FIB: Focused Ion Beam; AFM: Atomic Force Microscopy; CL: Cathodoluminescence; FWH<: full width at half-maximum; RMS: Root-Mean-Square; SEM: Scanning Electron Microscope; GL: Green Luminescence; BL: Blue Luminescence; DAP: Donor-Acceptor Pair.

Introduction

Driven by its potential application on high power electronics and solar-blind ultraviolet (UV) photodetectors

(PDs), gallium oxide in its most thermodynamically stable monoclinic structure, β-Ga_2O_3, gradually enters people's field of vision. Compared to GaN and SiC, β-Ga_2O_3 has a wide band gap (4.3-4.9 eV), high breakdown electric field (8 MV/cm) and excellent Baliga's figure (3214) [1-5]. Therefore, β-Ga_2O_3 is attracting interest for solar-blind self-powered UV PDs [6-11], Schottky barrier diodes (SBDs) [12], metal-semiconductor field-effect-transistors (MESFETs) [2] and metal-oxide-semiconductor field-effect-transistors (MOSFETs) [13-15].

The possibility of depositing β-Ga_2O_3 films on native substrates grown from the melt can allow synthesis of high-quality films and large-scale production, nonetheless, the high price hinders its possible application. Hence, heteroepitaxial β-Ga_2O_3 films have been obtained on several substrates, such as α-Al_2O_3 (0001) [16,17], MgO (100) [18], GaN (0001) [19,20], STO (100) [21] and $KTaO_3$ (100) [22]. At the same time, the corresponding photodetectors have been prepared and achieved excellent performance [23]. Among them, Guo, et al. [9,10] prepared a heterojunction by depositing n-type β-Ga_2O_3 thin film on p-type GaN by pulse laser deposition (PLD) to realize a super-high-performance self-powered UV photodetector, which helped solve the energy issues. However, the β-Ga_2O_3 thin film grown on GaN by molecular beam epitaxy (MBE) have been investigated to date seldom.

Recently, Nakagomi, et al. [20] reported that the orientation of β-Ga_2O_3 thin film formed on GaN template substrate was found to be (-201) β-Ga_2O_3 || (0001) GaN || (0001) sapphire and (010) β-Ga_2O_3 || {11-20} GaN, resulting in six-fold domain structure of the β-Ga_2O_3 layer. The β-Ga_2O_3 film grown on GaN substrate by metal organic chemical vapor deposition (MOCVD) is amorphous and transformed into (100) crystalline phase by annealing in oxygen atmosphere [24]. Li, et al. [19] reported the growth of vertical β-Ga_2O_3 nanowire arrays on GaN layers by MOCVD. The effect of the growth temperatures on the β-Ga_2O_3 thin film formed on GaN substrate remained unexplored, which is one of the crucial parameters for achieving high crystalline quality. In this work, the influence of substrate temperatures on the film quality is studied by using the ozone MBE. In addition, the epitaxial relationships, morphology and the optical properties are analyzed.

Experiments

The β-Ga_2O_3 thin films on (0001) Ga-plane GaN template (4 μm on (0001) sapphire substrate) were deposited by a commercial MBE (Octoplus-O 400, Komponenten) with liquid Ga (99.9999%) and ozone as the Ga source and O source. The GaN template substrate was cleaned by ultrasonic agitation in acetone, isopropyl alcohol and deionized water for 15min each, followed by drying with nitrogen gas blowing. During the growth, the ozone pressure of growth chamber was maintained at 5×10^{-6}mbar, and the Ga flux was 0.14Å/s which was detected by a quartz monitor crystal. The substrates were heated to growth temperature (500°C ~ 700°C), and then growth for 3h.

X-ray diffraction (XRD; D8 Advanced, Bruker) was carried out to check out the crystalline orientations and crystal quality. The film thickness was determined by cross-section observation with a focused ion beam (FIB; Scios, FEI), meanwhile, the surface morphology of the film was obtained from the plane view. The surface roughness of the film was analyzed by atomic force microscopy (AFM; Asylum Research MFP-3D, Oxford). The optical properties of β-Ga_2O_3 films was characterized by scanning cathodoluminescence (CL; Delmic Sparc, FEI).

Results and Discussion

The XRD θ-2θ scans of β-Ga_2O_3 thin films prepared with different substrate temperatures are shown in Figure 1(a). After the deposition of β-Ga_2O_3 thin films with the temperature changed from 500 to 700 °C, three diffraction peaks appear and locate at 18.96°, 38.36° and 58.09°, respectively, which correspond to the (-201), (-402) and (-603) lattice planes of monoclinic β-Ga_2O_3 (PDF# 43-1012). This indicates that the (-201) planes in the β-Ga_2O_3 thin film are parallel to the (0001) GaN surface. In addition, when the substrate temperature is 500 °C, the unwanted plane of (-801) is shown in the XRD θ-2θ scan, demonstrating the polycrystalline nature of this film. As the increase of substrate temperature, the (-801) plane disappears, indicating that when the substrate temperature above 550 °C, the β-Ga_2O_3 thin film is grown with the single orientation along (-201) lattice plane on the GaN template substrate. Figure 1(b) shows the XRD rocking curves of the (-201) plane of the β-Ga_2O_3 thin films prepared at different growth temperatures. And the full width at half-maximum (FWHM) values of rocking curves are plotted as a function of the substrate temperature in Figure 1(c). Apparently, the FWHM value monotonically decreases with increasing growth temperature and down to 1.67° at 700 °C, implying excellent crystal quality and unique (-201) out-plane orientation of the thin film.

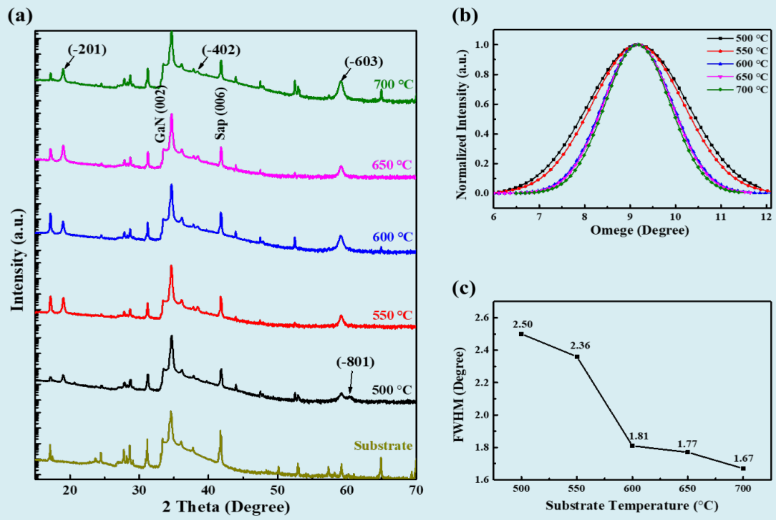

Figure 1: (a) θ-2θ XRD patterns of β-Ga_2O_3 thin films deposited on GaN template at various substrate temperatures. (b) XRD rocking curves of the (-201) plane of the β-Ga_2O_3 films and (c) the dependence of rocking curves FWHM values on the substrate temperature.

Figure 2(a) shows the XRD Φ-scan results for the β-Ga_2O_3 {-401} diffraction of the film grown at 700 °C. It can be seen that there are 6 diffraction peaks, derived from the β-Ga_2O_3 {-401} diffraction with the rotation angle offset 30°. Furthermore the 6 peaks appear every 60° are observed from the six peaks which associated with the {11-22} diffractions of GaN. This result indicates that the β-Ga_2O_3 thin film has a six-fold domain structure and an epitaxial relationship with the GaN. The schematic diagram of epitaxial relationship is presented in Figure 2(b). Base on it one can get that epitaxial relationships between (-201) plane of β-Ga_2O_3 (orange) and (0001) plane of GaN (blue) are β-Ga_2O_3 [010] // GaN [11-20] and β-Ga_2O_3 [102] // GaN [1-100]. The appearance of six-fold domain structure is due to the three-fold rotation symmetry corresponds to the epitaxial relationship and the originally two-fold β-Ga_2O_3 epitaxial growth in the three different directions at same rates [20].

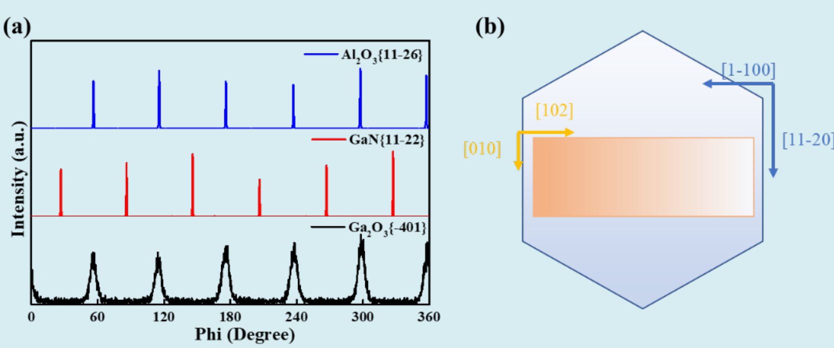

Figure 2: (a) XRD Φ-scan patterns for (top) {11-26} planes of Al_2O_3 substrate, (middle) {11-22} planes of GaN template and (bottom) {-401} planes of β-Ga_2O_3 film grown at 700°C. (b) Schematic diagram of epitaxial relationship between β-Ga_2O_3 (-201) plane (orange) and GaN (0001) plane (blue).

Figure 3 shows the cross-sectional scanning electron microscope (SEM) images of the β-Ga_2O_3 films prepared with different substrate temperatures and fabricated by FIB. The carbon layer and platinum layer are covered on the surface to are used to protect the surface before FIB. It can be seen that the thickness of film is stable around 144 nm. However, the film thickness decreases to 112 nm with the substrate temperature set at 700 °C. This may be due to the enhanced of the desorption or evaporation rate of volatile Ga_2O suboxide from the substrate when the temperature exceeds 700 °C [16,25].

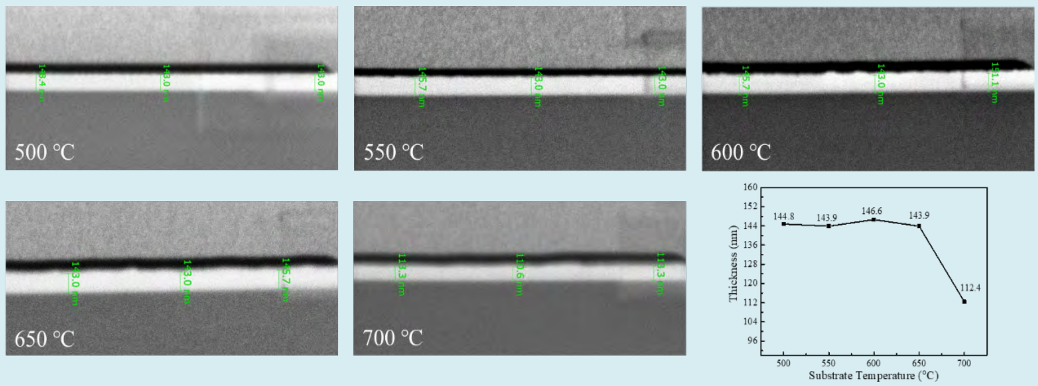

Figure 3: The cross-sectional SEM images of the β-Ga_2O_3 films deposited at different substrate temperatures. And the thickness of β-Ga_2O_3 films as a function of the substrate temperature.

The SEM plane-view images of β-Ga_2O_3 thin films grown at various temperatures are shown in Figure 4. It can be clearly observed that the β-Ga_2O_3 thin film grown on GaN template substrate follows the island-growth model, and the island size of β-Ga_2O_3 thin film increases as the substrate temperature increases. We believe that the island size difference is ascribed to the growth mechanism with different substrate temperature. When the substrate temperature is fixed at 500 °C, the adatoms do not have enough mobility, which limits the atomic migration distance and determine the size of the island. Thus, the polycrystalline thin film forms, which is consisted with the XRD results as shown in Figure 1(a). It is notable that the surface prefers a low free energy, the appeared morphology should be spherical shaped island, if the substrate structure is not similar to that of the epitaxial film. Hence, considering the influence of the substrate lattice structure, island structure appears as has been observed in the SEM images. Upon increasing the substrate temperature, the increased atomic mobility causes the radius of island to become larger, and nearby island begins to join together as shown in the SEM images.

Figure 5 displays the AFM morphologies and roughness of β-Ga_2O_3 thin films. The change of morphology is consistent with the SEM images with the substrate temperature increase. Corresponding to the changes of the surface morphology, the root-mean-square (RMS) roughness of the films increases when the substrate temperature does not exceed 650 °C. The maximum roughness of the films is 6.15 nm, indicating smooth surface.

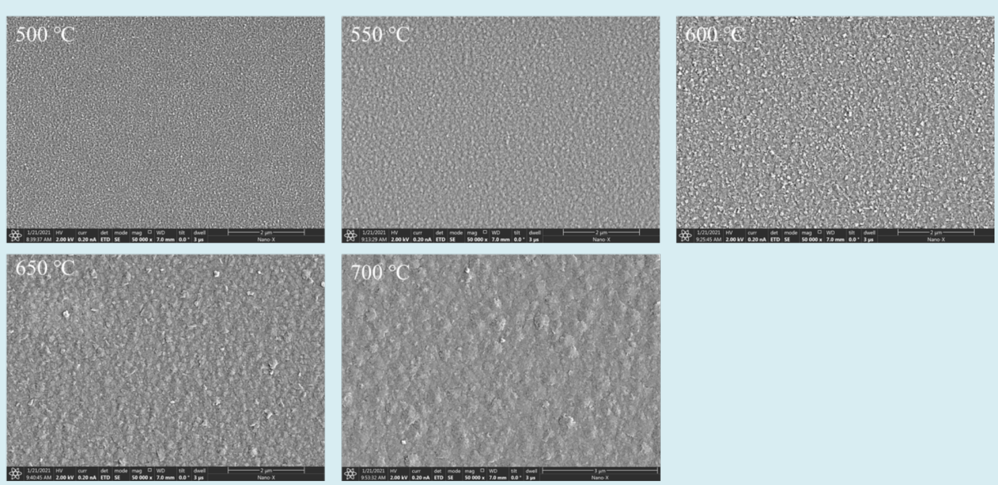

Figure 4: SEM plane-view images of the β-Ga_2O_3 films deposited at various temperatures.

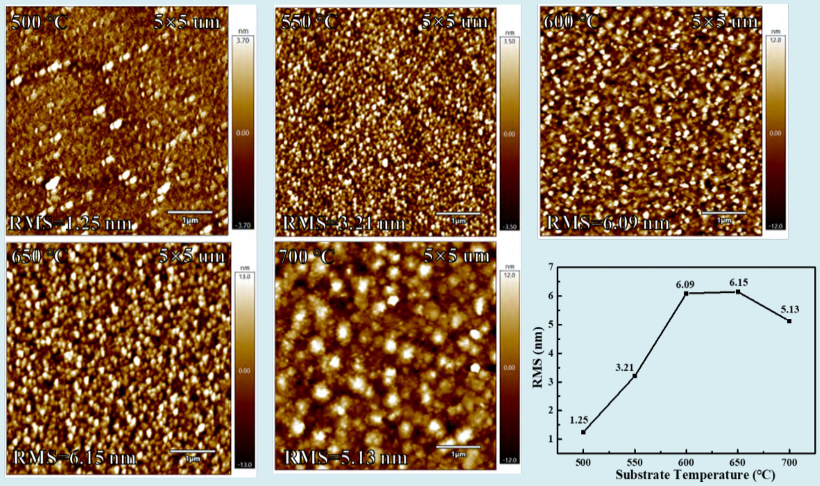

Figure 5: AFM images of the surface morphology for β-Ga_2O_3 films deposited at 500, 550, 600, 650 and 700 °C. And the RMS values of surface roughness of β-Ga_2O_3 films as a function of substrate temperature.

The room-temperature CL results from the β-Ga_2O_3 thin films are found to be strongly dependent on the substrate temperatures as shown in Figure 6. Three major light emission features are obvious from each spectrum and the energy correspond to 2.4 eV, 2.8-3.0 eV and 3.2-3.6 eV, named as green luminescence (GL) band, blue luminescence (BL) band and UV band, respectively. The UV band is generally assigned to recombination of free electrons and self-trapped holes [26-28]. And the BL band may result from defect related luminescence, attributed to donor-acceptor pair (DAP) recombination. Possible donors are intrinsic point defects such as oxygen vacancies (V_o) and interstitial Ga (Ga_i), and possible acceptors are Ga vacancies (V_{Ga}), V_o-V_{Ga} complexes [26-28]. Thus, the change of CL spectra is considered to be caused by the variation of crystal quality of the β-Ga_2O_3 thin films. As the substrate temperature increases, the luminescence of BL band and GL band gradually decreases, while the UV band luminescence increases, illustrating the improvement of crystal quality. In order to further quantitatively compare the luminescence ratio, each spectrum is fitted by four peaks, as shown in Figure 7(a)-(e). Note that the UV is fitted by the two peaks because the photogenerated holes can self-trap onto two different O sites [29,30]. Figure 7f shows the summation area of BL an GL peaks with respect to the area of the UV peak as a function of the substrate temperature. The relative intensity ratio of BL and GL to UV decreases with the increasing of substrate temperature, and the ratio reaches the lowest value when the substrate temperature is 700°C. This result implies that among these substrate temperatures, the β-Ga_2O_3 thin film has the best crystal quality at the substrate temperature of 700°C, which is consistent with the XRD rocking curves as shown in Figure 1b.

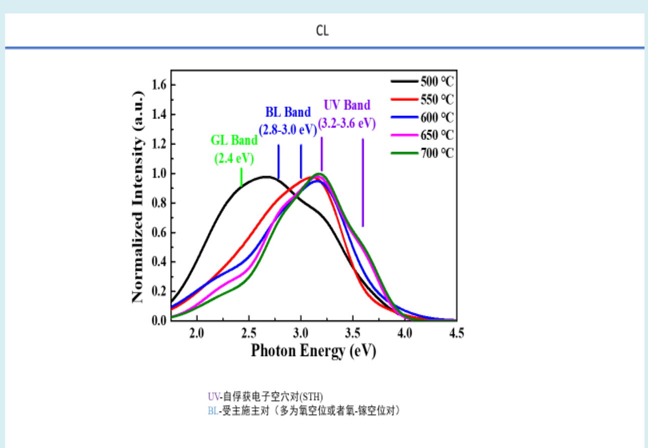

Figure 6: CL spectra of β-Ga_2O_3 films grown at different substrate temperatures.

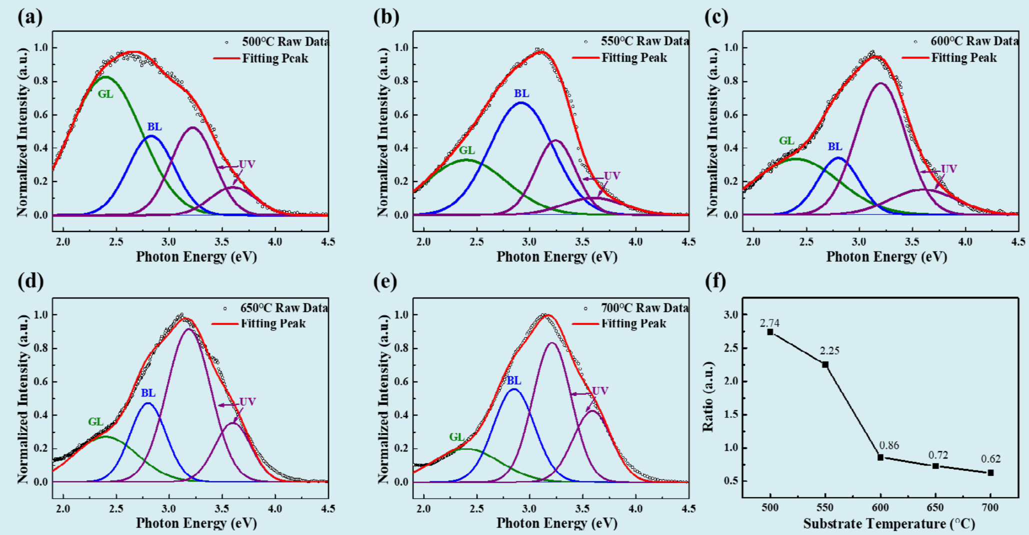

Figure 7: (a)-(e) CL spectra with curve-fitting results of β-Ga_2O_3 thin films grown at different substrate temperatures. (f) The dependence of the ratio (the summation area of BL and GL peaks to the area of UV peaks) on the substrate temperature.

Conclusion

In conclusion, β-Ga_2O_3 thin films are grown on c-plane GaN template substrate by ozone MBE. Phase-pure (-201)-oriented β-Ga_2O_3 thin film can be formed with the substrate temperature exceeded 550 °C, and the crystal quality improves with increasing substrate temperature. The epitaxial relationships are confirmed as β-Ga_2O_3 [010] // GaN [11-20] and β-Ga_2O_3 [102] // GaN [1-100], and result six-fold domain structure in β-Ga_2O_3 thin film. Because the difference in crystal structure between the film and the substrate, the β-Ga_2O_3 thin film follows the island-growth model, and the size of the island increases as the substrate temperature increases. In addition, CL spectra variations are observed that the defect related luminescence decreases with the increase of substrate temperature, claiming the improvement of crystal quality which are consistent with the results of XRD rocking curves. The presented optimized β-Ga_2O_3 thin film grown on GaN template substrate should effectively promote development of reliable high performance self-powered UV photodetector based on the Ga_2O_3/GaN heterojunction.

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