Metalorganic Chemical Vapor Deposition of (100) β-Ga_2O_3 on on-axis Ga_2O_3 substrates

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Abstract

MOCVD growths of β-Ga_2O_3 on on-axis (100) Ga_2O_3 substrates are comprehensively investigated. Key MOCVD growth parameters including growth temperature, pressure, group VI/III molar flow rate ratio, and carrier gas flow rate are mapped. The dependence of the growth conditions is correlated with surface morphology, growth rate and electron transport properties of the MOCVD grown (100) β-Ga_2O_3 thin films. Lower shroud gas (Argon) flow is found to enhance the surface smoothness with higher room temperature (RT) electron Hall mobility. The growth rate of the films decreases but with an increase of electron mobility as the VI/III molar flow rate ratio increases. Although no significant variation on the surface morphologies is observed at different growth temperatures, the general trend of electron Hall mobilities are found to increase with increasing growth temperature. The growth rates reduce significantly with uniform surface morphologies as the chamber pressure increases. By tuning the silane flow rate, the controllable carrier concentration of (100) β-Ga_2O_3 thin films between low-10^{17} cm^{-3} to low-10^{18} cm^{-3} was achieved. Under optimized growth condition, an (100) β-Ga_2O_3 thin film with RMS roughness value of 1.64 nm and a RT mobility of 24 cm^2/Vs at a carrier concentration of 7.0 x 10^{17} cm^{-3} is

demonstrated. The mobilities are primarily limited by the twin lamellae and stacking faults defects generated from the growth interface. Atomic resolution scanning transmission electron microscopy (STEM) reveals the formation of twin boundary defects in the films, resulting in the degradation of crystalline quality. Results from this work provide fundamental understanding of the MOCVD epitaxy of (100) β-Ga_2O_3 on on-axis Ga_2O_3 substrates and the dependence of the material properties on growth conditions. The limitation of electron transport properties of the (100) β-Ga_2O_3 thin films below 25 cm^2/Vs is attributed to the formation of incoherent boundaries (twin lamellae) and stacking faults grown along on-axis (100) crystal orientation.

Keywords: Ultrawide bandgap semiconductor, β-Ga_2O_3 thin films, (100) orientation, Si doping, metalorganic chemical vapor deposition

I. Introduction

Monoclinic β-Ga_2O_3 is an emerging semiconductor material with an ultrawide bandgap of ~4.8 eV and high predicted breakdown field strength of 8 MV/cm [1, 2]. Due to its high Baliga's figure of merit, thermal and chemical stability, controllable n-type doping with excellent transport properties and its ability of bandgap engineering by alloying with Al_2O_3 or In_2O_3 [3-23], β-Ga_2O_3 is considered as a promising candidate for next generation high power and radio frequency electronics. Another key advantage of β-Ga_2O_3, in comparison to other wide bandgap materials (e.g., GaN, SiC) or ultrawide bandgap materials (e.g., diamond, AlN) is its availability of single crystal high quality native substrates with different orientations grown by scalable melt growth methods [24-27]. Several β-Ga_2O_3-based electronic and optoelectronic devices such as lateral [2,28,29] and vertical field-effect transistors [30,31], high-breakdown Schottky barrier diodes [32-34], and solar-blind ultraviolet photodetectors [35] have already been successfully demonstrated

on β-Ga_2O_3 films grown on native substrates, which reveals a great potential of this material for future device technologies.

Epitaxial growth of high-quality thin films on β-Ga_2O_3 native substrates is crucial for the development of high-performance devices. Efforts on the homoepitaxial growth of β-Ga_2O_3 thin films have been made via different growth methods such as metalorganic chemical vapor deposition (MOCVD) [3,4,6,8,10,36], molecular beam epitaxy (MBE) [5,37,38], low pressure chemical vapor deposition (LPCVD) [39-41], and halide vapor phase epitaxy (HVPE) [42-45] on different orientations of β-Ga_2O_3 native substrates including (100), (010), (001), and ($\bar{2}$01). The crystal orientation of β-Ga_2O_3 substrates plays an important role that significantly influence the quality of the epi-films. In recent years, outstanding results from the homoepitaxial growth of (010) β-Ga_2O_3 thin films have been demonstrated by MOCVD, which include controllable n-type doping over a wide range of electron density (10^{16} - 10^{19} cm^{-3}) with high room (~200 $cm^2/V.s$) and low temperature (>10^4 $cm^2/V{\cdot}s$) electron mobilities, approaching the theoretical limits with ultra-low background concentrations (~10^{14} cm^{-3}) [3,4,6,8,10]. On the other hand, higher growth rates of β-Ga_2O_3 have been demonstrated by HVPE and LPCVD growth methods [40,42-45]. Additionally, the growth rate on (010) oriented substrates via MBE was found to be significantly higher than the (100) orientation [46]. As compared to other orientations, (100) plane of β-Ga_2O_3 is the preferred cleavage plane with the lowest surface energy and can easily be prepared [47]. Step flow homoepitaxial growth of β-Ga_2O_3 thin films on (100) oriented off-axis β-Ga_2O_3 substrates with miscut angles have been demonstrated with superior crystalline quality [36]. While decent electron transport results have been achieved for the growth on (100) plane by introducing appropriate substrate miscut angles (6°), β-Ga_2O_3 films grown on on-axis (100) plane suffer from high density of twins and stacking faults [48-52].

Although the homoepitaxial growth of β-Ga_2O_3 films grown on on-axis (100) plane of β-Ga_2O_3 substrates were characterized by 2D island growth mode with the formation of incoherent boundaries (twin lamellae), recent MOCVD growth of β-$(Al_xGa_{1-x})_2O_3$ thin films on on-axis (100) plane exhibited high-Al incorporation up to $x \leq 52\%$ without occurring phase segregation [15], indicating great promises for bandgap engineering on (100) oriented substrates. In addition, as compared to other orientations, the highest conduction band offsets are experimentally measured at (100) β-Ga_2O_3/β-$(Al_xGa_{1-x})_2O_3$ heterointerfaces [16,22,53], showing great promise for electron confinement at the interface for the development of modulation doped field effect transistors (MODFETs) with high mobility. High Al incorporation in (100) β-$(Al_xGa_{1-x})_2O_3$ films was found to promote the step flow growth with smooth and uniform surface morphology where Al adatoms act as preferential nucleation sites during the growth [15]. To improve the structural quality of the material and to find optimum growth conditions, the basic understanding of the influence of MOCVD growth parameters on the electrical properties of β-Ga_2O_3 films grown on (100) on-axis β-Ga_2O_3 substrates is necessary.

In this work, we have investigated the impact of key MOCVD growth parameters such as the growth temperature, chamber pressure, VI/III ratio, shroud (carrier) and silane (SiH_4) gas flow rates, on the structural, electrical, and surface morphological properties of β-Ga_2O_3 thin films grown on (100) β-Ga_2O_3 substrates. The systematic growth mapping and comprehensive material characterization via high resolution scanning transmission electron microscopy (HR-STEM) scanning electron microscopy (SEM), atomic force microscopy (AFM), x-ray diffraction (XRD) and room temperature Hall measurements were performed to investigate the growth window for achieving better crystalline quality of β-Ga_2O_3 epitaxial films on (100) Ga_2O_3 substrate.

II. Experimental Section

The β-Ga_2O_3 films were grown via MOCVD on Fe-doped semi-insulating (100) β-Ga_2O_3 substrates without intentional miscut angle. The substrates were commercially acquired from Novel Crystal Technology, Inc. Si-doped β-Ga_2O_3 films with thickness ranging from ~280-860 nm were grown for 1 hour by varying the growth temperature from 650 to 950 °C, and chamber pressure from 20 to 100 torr. Triethylgallium (TEGa) and pure O_2 were used as Ga and O precursors, respectively. Ar was used as carrier gas. O_2 flow rate was fixed at 800 sccm. Silane flow rate was tuned between 0.227 and 44.64 nmol/min. Shroud (Ar) gas flow was tuned from 300 to 2800 sccm. Different VI/III molar flow ratio of 1150, 1495, 2135 and 2989 were obtained by tuning the TEGa flow from 130 to 50 sccm and using a constant O_2 flow rate of 800 sccm. The influence of the chamber pressure was studied for two growth temperatures of 700 and 880 °C. Prior to loading the substrate in MOCVD chamber, the substrates were ex-situ solvent cleaned. High temperature (920 °C) in-situ annealing was performed for 5 mins under O_2 atmosphere for removing any potential contaminants from the substrate surface. The list of growth parameters and film thicknesses with corresponding sample IDs are summarized in Table 1 and 2.

Room temperature Hall measurements using Ecopia HMS-3000 Hall effect system were performed to measure the carrier concentrations and mobilities at room temperature, with a fixed magnetic field of 0.975 T. In order to create the van der Pauw geometry for the Hall measurement, Ti/Au (30/100 nm) contacts were deposited on the four corners of the sample to form ohmic contacts. Surface morphologies were characterized by field emission scanning electron microscopy (FESEM) by using Nanolab 600. The film thicknesses and growth rates were estimated by examining the cross sectional FESEM images of the Ga_2O_3 films grown on the coloaded sapphire substrates. The surface roughness was acquired from AFM using a Bruker Icon

3. The crystalline quality β-Ga_2O_3 films were probed by x-ray rocking curve (XRC, Bruker D8 Discover). A Thermo Fisher Scientific Themis-Z scanning transmission electron microscope (operated at 200 kV) was used for high resolution STEM imaging.

III. Results and Discussions

The influence of shroud gas (Ar) flow rates on the surface morphology and electron transport properties of β-Ga_2O_3 films are investigated by varying the Ar flow rates from 100 to 2800 sccm, as listed in Table 1 (Sample ID: GE04, GE11, GE10, GE09, GE12). The other parameters were kept as constants (VI/III ratio at 2135, growth temperature at 880 °C and chamber pressure at 60 torr). The surface morphology of the β-Ga_2O_3 films grown with different shroud gas flow were evaluated by FESEM and AFM imaging. Figures 1(a)-(e) show the surface FESEM images of β-Ga_2O_3 films grown at shroud gas flow rates of 100-2800 sccm, respectively. Although all the surfaces show the appearance of bump like 3D island structures, the size and density of these structures are found to decrease as the shroud gas flow decreases. Smoother surface morphology is observed for the films grown with 100 and 300 sccm flow rates, indicating that the lower shroud gas flow can promote surface smoothening. The corresponding surface AFM images (over a scan area of 5 x 5 μm^2) for the films are shown in Figures 2(a)-(e). The size of the 3D structures as indicated by the white spots in the AFM images reduces with the decrease of shroud gas flow. Figure 3(a) plots the surface RMS roughness values of the films as a function of different shroud gas flow rates. The RMS roughness decreases monotonically from 12.1 to 6.29 nm as the shroud gas flow decreases from 2800 to 100 sccm, which is consistent with the smoother surface morphology as observed in FESEM images in Figure 1(a)-(b) for the samples grown with lower shroud gas flows (100/300 sccm).

The growth rates are also found to be highly dependent on the shroud gas flows as shown in Figure 3 (b). While almost similar growth rates (~850 nm/h) are observed for < 1100 sccm of flows, further increase of the flow rates from 1100 to 2800 sccm results in the decrease of growth rates to 617 nm/h, which can primarily be attributed to the decrease of the partial pressure of the Ga and O precursors due to higher shroud gas flows. It is also worth noting that the flow pattern in the reactor could greatly alter the amount of chemical species transported to the growth surface.

The room temperature (RT) Hall mobility is also affected by different shroud gas flows as shown in Figure 3 (c). The carrier concentrations of the films grown with different shroud gas flow rates varied from 2.0×10^{17}- 4.3×10^{17} cm^{-3}. Relatively higher mobility is obtained for lower shroud gas flow. The RT mobility of 11 $cm^2/V.s$ with carrier concentration of 2.3×10^{17} cm^{-3} is measured at 300 sccm shroud gas flow. As the shroud gas flow rate increases, the partial pressure of O and Ga decreases, which in turn influences the amount of chemical species transported to the growth surface and impact the crystalline quality of the film. As a result, the Hall mobility decreases, indicating the degradation of crystalline quality, which is also evidenced by the rougher surface morphology of the films grown with higher shroud gas flow (Figure 2(e)).

The surface morphology, growth rates and transport properties of β-Ga_2O_3 films are also systematically investigated as a function of VI/III molar flow ratio. The VI/III ratio was varied from 1150 to 2989, by tuning the TEGa flow from 31.06 to 11.95 μmole/min (Sample ID: GE09, GE14, GE13, GE15 in Table 1). The other parameters such as the O_2 (800 sccm) and shroud gas flows (300 sccm), growth temperature (880 °C) and chamber pressure (60 torr) were kept constant. The surface FESEM images of β-Ga_2O_3 films grown with VI/III ratio ranging from 1150 to 2989 are shown in Figures 4(a)-(d), respectively. The surface becomes uniform and smoother with smaller and less dense 3D island structures, as the VI/III ratio increases from 1150 to 2135. Further

increase of the VI/III ratio leads to rougher surface morphologies with higher density of bumps. The smoother surface morphology with relatively smaller islands is observed for VI/III ratio of 2135 as shown in Figure 4(c). The corresponding surface AFM images as shown in Figures 5(a)-(d), also indicate the presence of 3D island structures on the surface of the films. The Surface smoothness enhances with lower RMS roughness (from 6.63 nm to 2.62 nm) as the VI/III ratio increases from 1150 to 2135, while further increase of the VI/III ratio to 2989 leads to higher RMS value of 3.06 nm (Figure 6(a)). Note that the observed trend in this study is different from that of (100) β-Ga_2O_3 grown on misoriented substrates [36]. Not only the different growth conditions including VI/III molar ratio, chamber pressure and growth temperature, but also the off-axis substrate can play important roles that affect the surface morphology of the grown films. In this study, the lowest RMS roughness of 2.62 nm is observed for VI/III = 2135, which is consistent with the smooth surface morphology of the films as shown in the SEM images of Figure 4(c). Note that under a different growth condition, the relatively optimized VI/III ratio for low RMS value is likely to be different.

We further investigated the dependence of the growth rates on TEGa molar flow rate as shown in Figure 6(c). The growth rates ranged between 380-860 nm/h depending on the TEGa molar flow rate from 11.95 to 31.06 μmole/min. A lower TEGa molar flow rate leads to the decrease of film growth rate. This can be attributed to the lower amount of chemical species (TEGa) that can reach to the growth surface under lower shroud gas flow of 300 sccm due to increased gas phase reaction.

The room temperature Hall mobility of β-Ga_2O_3 films is also investigated for different VI/III ratio as shown in Figure 6 (b). Hall mobilities ranging between 7-11 $cm^2/V.s$ are measured with 2.3 x 10^{17}- 4.7 x 10^{17} cm^{-3} doping concentrations for VI/III ratio of 1150-2135. However, the mobility of the film grown at VI/III ratio of 2989 cannot be accurately measured, which was due

to the strong compensation from the formation of defects (e.g, incoherent twin boundaries) at relatively high VI/III ratio, indicating strong influence of VI/III ratio on the transport properties of the β-Ga_2O_3 thin films.

Silane flow rate is tuned from 0.23 to 44.64 nmol/min to investigate the effect on carrier concentration and mobility of β-Ga_2O_3 films (Sample ID: GE17, GE13, GE18, GE24, GE19 in Table 1). The other parameters such as the shroud gas flow of 300 sccm, VI/III ratio of 2135, growth temperature of 880 °C and chamber pressure of 60 torr were selected based on the relatively smooth surface morphology and better mobility of the films, as discussed in the previous sections. The room temperature Hall mobility and carrier concentration as a function of different silane flow rates are shown in Figure 7. By tuning the silane flow rates from 0.23 nmol/min to 44.64 nmol/min, the carrier concentrations ranging from ~$3.2x10^{17}$ cm^{-3} to ~$7.0x10^{17}$ cm^{-3} are achieved. The highest RT mobility of ~24 $cm^2/V.s$ is measured at the carrier concentration of ~$7.0x10^{17}$ cm^{-3}. The relatively low carrier concentrations observed at high silane flow rates can be due to the strong compensation from the twin boundary defects formed on (100) on-axis growth surface plane.

The growth temperature was varied from 650 to 950 °C to investigate its effect on the growth of (100) β-Ga_2O_3 films (Sample ID: GE27, GE26, GE25, GE18, GE30 in Table 1). The optimized shroud gas flow of 300 sccm, VI/III ratio of 2135 and chamber pressure of 60 torr were used for this study. No significant effect of the growth temperature on the surface morphology is observed from the FESEM images as shown in Figures 8 (a)-(e). All the surfaces exhibit uniform and smooth morphologies. The corresponding surface AFM images as shown in Figures 9 (a)-(e) also indicate similar RMS roughness values ranging between 1.40 and 1.64 nm although the growth temperatures are varied for a wide range (650-950 °C), indicating minimum influence of growth temperature on the surface morphologies of the films.

While no noticeable impact on the surface morphologies is observed for different growth temperature, the growth rates of β-Ga_2O_3 films on (100) plane are found to be significantly influenced as shown in Figure 10 (b). With the increase of growth temperature from 650 to 800 °C, the growth rate decreases from 581 to 440 nm/h which can be due to higher desorption of Ga adatoms from the growth surface as temperature increases. However, further increase of the temperature to 950 °C increases the growth rate to 528 nm/h, which can be attributed to the higher surface reaction rates of Ga adatoms on growth surface at elevated growth temperatures.

The transport properties are also investigated as a function of growth temperature as shown in Figure 10 (c). The carrier concentrations vary between 7 x 10^{17} and 1.8 x 10^{18} cm^{-3} as the temperature is tuned from 650 to 950 °C. The highest mobility of 24 $cm^2/V.s$ at carrier concentration of ~7.0x10^{17} cm^{-3} was measured at 880 °C. The general trend shows that the mobility increases with the increase of growth temperature, indicating strong influence of growth temperature on Hall mobility of β-Ga_2O_3 films. A higher growth temperature reduces the impurity incorporation, and promotes the surface diffusion of adatoms, which in turn increases the epi-film quality with higher mobility.

The influence of chamber pressure ranging from 20-100 torr is also evaluated by comprehensive characterization of SEM, AFM and Hall measurements for two different growth temperatures of 700 and 880 °C. The sample IDs (GE26, GE28, GE29, GE31, GE32 and GE33) with corresponding film thicknesses are listed in Tables 1 and 2. The films were grown with the shroud gas flow of 300 sccm and VI/III ratio of 2135 for 1 hr growth duration. The growth rates, as listed in Table 2 decreases significantly with the increase of chamber pressure for both 700 and 880 °C growth temperature, indicating strong impact of growth pressure on the growth rates of MOCVD grown (100) β-Ga_2O_3 thin films. This can be attributed to the increase of the gas phase

reaction of precursors at relatively higher chamber pressure. Similar trend of reduction of growth rates at higher chamber pressure was also observed previously for MOCVD growth of (010) oriented β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$ thin films [3,18].

The surface morphology of the β-Ga_2O_3 films grown with different pressures was evaluated by FESEM and AFM imaging. The surface FESEM images of β-Ga_2O_3 films grown with chamber pressures of 20-100 torr are shown in Figures 11 (a)-(c) and Figures 12 (a)-(c) for 700 °C and 880 °C growth temperatures, respectively. While noticeable 3D island structures are observed at 20 torr chamber pressure for both 700 °C (Figure 11(a)) and 880 °C (Figure 12(a)) temperatures, higher chamber pressures (60 and 100 torr) are found to result in featureless surface morphology with enhanced uniformity (Figures 11(b-c) and 12(b-c)). The surface features as characterized by 3D island growths under low pressure (20 torr) could be due to the significant larger diffusion length of adatoms on the growth surface. However, smoother surface morphologies observed at higher chamber pressure can also be due to the reduced film thicknesses. The surface AFM images in Figures 11 (d)-(f) and Figures 12 (d)-(f) show a similar trend of smoother surface morphologies at higher pressure. As compared to the variation of surface RMS values (3.47 to 1.54 nm) of the films grown at 700 °C, obvious reduction of the RMS values from 20 to 1.18 nm is observed for the films grown at 880 °C as the chamber pressure increases from 20 to 100 torr, indicating strong impact of the chamber pressure on the surface morphologies of β-Ga_2O_3 films grown at higher growth temperature.

The room temperature Hall mobility and carrier concentrations of β-Ga_2O_3 films grown at different chamber pressures is also investigated as listed in Table 2. The carrier concentrations are strongly influenced by the chamber pressure. With the same silane flow of 8.29 nmole/min, the carrier concentration decreases from 1.8×10^{18} to 1.7×10^{17} cm^{-3} as the chamber pressure increases

from 60 to 100 torr at 700 °C growth temperature. Similarly, at growth temperature of 880°C, the carrier concentrations reduce monotonically from 9.0 x 10^{17} to 2.4 x 10^{17} cm^{-3} as the pressure increases from 20 to 100 torr, which can be attributed to the lower growth rates of β-Ga_2O_3 films at higher chamber pressure. While the films grown at 700 °C exhibited lower mobilities, relatively higher RT mobility of 23 $cm^2/V.s$ at 7.4 x 10^{17} cm^{-3} carrier concentration was measured at growth temperature of 880 °C and chamber pressure of 60 torr.

To investigate the effect of growth conditions on crystalline quality, scanning transmission electron microscopy (STEM) characterization was performed on Ga_2O_3 films grown at different growth rates. Figure 13 (a)-(b) shows the STEM high-angle annular dark-field (HAADF) and low-angle ADF (LAADF) STEM images of the sample grown by an initial condition (sample GE04). The inset represents the cross-sectional viewing direction of [010] for the β-Ga_2O_3 crystal structure. The STEM results show the layer is barely distinguishable from the substrate, indicating defect-free interfaces between the substrate and the epi-layers. Moreover, there are horizontal band contrasts (Figure 13(b)), revealing the extended defects formed in these epitaxial layer regions. Figure 13(c) shows that a crystallographic translation was observed at the interface. A mirror operation of the β-Ga_2O_3 crystal structure along the (100) crystal plane, and a half c-lattice transition along the [100] direction was exhibited during the epitaxial growth of the β-Ga_2O_3 layer. In addition, it is noted that the horizonal band contrasts in a magnified view of region (Figure 13(d)) were characterized as stacking mismatch boundaries lying on the (001) crystal plane. Two coherent boundaries where the upper sheet is a half c-lattice shifted and overlaid to another layer, resulting in a defect structure with superimposed Ga atoms. This type of extended defect resembles to the atomic structure which contains the formation of twin lamellae through double positioning occurred in the step flow growth mechanism [51].

On the other hand, Figure 14 displays the STEM results of sample GE18 grown by an optimized growth condition. STEM HAADF images show a defect-free region at the interface (Figure 14(a)). The horizontal band contrasts are significantly lower in the film region as compared to the sample GE04 (Figure 14(b)), while the vertical contrast represents a thickness variation of the TEM foil. Figure 14(c) shows a magnified view at the interface, indicating a similar lattice translation sequence and stacking throughout the layer. Figure 14(d) reveals the stacking mismatch boundaries overlaid to each other, resulting in the defect structure with the superimposed cation columns. The stacking mismatch boundaries extend along the c-direction and proceed through the entire layer to the surface, reducing the growth quality of epitaxial β-Ga_2O_3 thin films significantly. For the given growth condition, reducing the density of extended defects is essential to enhance the Hall mobility of epitaxial (100) β-Ga_2O_3.

Figure 15 shows the XRD rocking curve profiles of the β-Ga_2O_3 films with different mobility (GE18 Mobility= 24 cm^2/Vs, GE30 Mobility= 17 cm^2/Vs and GE11 Mobility= 4 cm^2/Vs). The sample GE18 shows the lowest rocking curve FWHM with the highest mobility, as compared to the other two samples with mobility of 17 and 4 cm^2/Vs and the corresponding rocking curve FWHM of 73 and 108 arcsec, respectively.

IV. Conclusions

In summary, systematic mapping of MOCVD growth of (100) β-Ga_2O_3 films is performed to investigate the structural, surface morphological and electrical properties of β-Ga_2O_3 films for a wide range of experimental growth window including the shroud gas (Ar) flow, growth temperature, reactor pressure, VI/III molar ratio and silane flow rates. The shroud (carrier) gas flow (Ar) and VI/III ratio have strong influence on the surface roughness. Decrease in shroud gas flow corresponding to the increase in the partial pressure of TEGa and oxygen can increase the

growth rate and surface smoothness of the films. The increase in the VI/III molar ratio reduces the growth rates with uniform and smooth surface morphology. Growth temperature has minimum effect on surface morphology, however it has more prominent impact on growth rate and mobility. In general, under high temperatures, the β-Ga_2O_3 films have higher mobility which are correlated to better crystalline quality. The growth pressure is found to have a significant impact on the surface morphology and growth rate. The increase of the pre-reactions of the precursors in gas phase at higher chamber pressure lowers the growth rate, which also leads to the smoother surface morphology. At high growth temperatures, the effect of growth pressure is more prominent. Results from this work will provide guidance for developing (100) β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$ films for future device technologies.

Conflict of Interest Statement

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Data Availability

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

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Table Captions

Table 1. Summary of (100) β-Ga_2O_3 films grown with different growth conditions: shroud gas flow rate (100-2800 SCCM), VI/III ratio (1150-2989), growth temperature (650-950 ℃), chamber pressure (20-100 torr), silane flow rate (0.227-44.64 nmol/min).

Table 2. List of (100) β-Ga_2O_3 films grown at different chamber pressure (20-100 torr) and temperature (700 ℃ and 880 ℃). The corresponding growth rate, charge concentration and room temperature electron Hall mobility are included.

Figure Captions

Figure 1. Surface view FESEM images of β-Ga_2O_3 films grown at different shroud gas flow rate: (a) 100 SCCM (GE12), (b) 300 SCCM (GE09), (c) 1100 SCCM (GE10), (d) 1900 SCCM (GE11), and (e) 2800 SCCM (GE04).

Figure 2. The surface AFM images (5µm x 5µm scan area) of β-Ga_2O_3 films grown at different shroud gas flow rate: (a) 100 SCCM (GE12), (b) 300 SCCM (GE09), (c) 1100 SCCM (GE10), (d) 1900 SCCM (GE11), and (e) 2800 SCCM (GE04).

Figure 3. The (a) RMS roughness, (b) growth rate, and (c) room temperature electron Hall mobility of β-Ga_2O_3 films as a function of the shroud gas flow rate (GE04, GE09, GE10, GE11, GE12).

Figure 4. Surface view FESEM images of β-Ga_2O_3 films grown with different group VI/III molar ratio: (a) 1150 (GE09), (b) 1495 (GE14), (c) 2135 (GE13), and (d) 2989 (GE15).

Figure 5. The surface AFM images (5µm x 5µm scan area) of β-Ga_2O_3 films grown with different group VI/III molar ratio: (a) 1150 (GE09), (b) 1495 (GE14), (c) 2135 (GE13), and (d) 2989 (GE15).

Figure 6. The (a) RMS roughness, (b) room temperature electron Hall mobility of β-Ga_2O_3 films as a function of the VI/III ratio, and (c) growth rate of β-Ga_2O_3 films as a function of the TEGa molar flow rate. (GE09, GE13, GE14, GE15).

Figure 7. The room temperature Hall mobilities and electron concentrations of β-Ga_2O_3 films as a function of the silane molar flow rate (GE13, GE17, GE18, GE19, GE24).

Figure 8. Surface view FESEM images of β-Ga_2O_3 films grown at different growth temperature: (a) 950 ℃ (GE30), (b) 880 ℃ (GE18), (c) 800 ℃ (GE25), (d) 700 ℃ (GE26), and (e) 650 ℃ (GE27).

Figure 9. The surface AFM images (5µm x 5µm scan area) of β-Ga_2O_3 films grown at different growth temperature: (a) 950 ℃ (GE30), (b) 880 ℃ (GE18), (c) 800 ℃ (GE25), (d) 700 ℃ (GE26), and (e) 650 ℃ (GE27).

Figure 10. The (a) RMS roughness, (b) growth rate, and (c) room temperature Hall mobility of β-Ga_2O_3 films as a function of the growth temperature (GE18, GE25, GE26, GE27, GE30).

Figure 11. Surface view FESEM images of β-Ga_2O_3 films grown at 700 ℃ at different chamber pressure: (a) 20 torr (GE28), (b) 60 torr (GE26) and (c) 100 torr (GE29). The corresponding surface AFM images with RMS roughness values are shown in (d-f).

Figure 12. Surface view FESEM images of β-Ga_2O_3 films grown at 880 ℃ at different chamber pressure: (a) 20 torr (GE31), (b) 60 torr (GE33) and (c) 100 torr (GE32). The corresponding surface AFM images with RMS roughness values are shown in (d-f).

Figure 13. Atomic resolution STEM images of the β-Ga_2O_3 film (GE04): (a) a STEM HAADF image, (b) a STEM LAADF image, (c) the interface region between the epi-layer and the substrate, and (d) a featured region in epitaxial layer.

Figure 14. Atomic resolution STEM images of the β-Ga_2O_3 film (GE18): (a) a STEM HAADF image, (b) a STEM LAADF image, (c) the interface region between the layer and the substrate, and (d) a featured region in the epitaxial layer.

Figure 15. XRC profiles of the β-Ga_2O_3 films with different mobilities (GE18 Mobility= 24 cm^2/V s, GE30 Mobility= 17 cm^2/V s and GE11 Mobility= 4 cm^2/V s).

Table 1

Sample ID	Shroud Gas (sccm)	VI/III ratio	Growth Temperature (°C)	Chamber Pressure (Torr)	Silane Flow (nmol/min)	Ga_2O_3 film thickness (nm)
GE04	2800	1150	880	60	2.212	617
GE11	1900	1150	880	60	2.212	740
GE10	1100	1150	880	60	2.212	865
GE09	300	1150	880	60	2.212	860.1
GE12	100	1150	880	60	2.212	842.3
GE14	300	1495	880	60	2.212	715.4
GE13	300	2135	880	60	2.212	553.2
GE15	300	2989	880	60	2.212	378.3
GE17	300	2135	880	60	0.227	485.9
GE18	300	2135	880	60	8.929	510
GE24	300	2135	880	60	25.51	575
GE19	300	2135	880	60	44.64	547.5
GE30	300	2135	950	60	8.929	528.1
GE25	300	2135	800	60	8.929	439.6
GE26	300	2135	700	60	8.929	470
GE27	300	2135	650	60	8.929	580.8
GE28	300	2135	700	20	8.929	625
GE26	300	2135	700	60	8.929	470
GE29	300	2135	700	100	8.929	320
GE31	300	2135	880	20	8.929	656
GE33	300	2135	880	60	8.929	500.1
GE32	300	2135	880	100	8.929	281

Table 2

Sample ID	Growth Temperature (℃)	Chamber Pressure (Torr)	Growth Rate (nm/hr)	Charge concentration (cm^{-3})	Hall mobility ($cm^2/V\ s$)
GE28	700	20	625	-	-
GE26	700	60	470	1.80E+18	4
GE29	700	100	320	1.70E+17	2
GE31	880	20	656	9.00E+17	4
GE33	880	60	500.1	7.40E+17	23
GE32	880	100	281	2.40E+17	4