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Investigation of Electron Transport in B-(Al, Ga)2O3 Thin Films and Heterostructures Under Applied High Pressures

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All samples were grown in a RIBER 32 hybrid MBE system equipped with conventional Ga and Ge thermal effusion cells. The oxygen source consisted of ultra-high purified oxygen (>99.999%) and was activated by the RIBER RF-O 50/63 oxygen RF plasma source. A plasma power and oxygen flow rate of 410W and 2sccm, respectively, and a Ga flux of 10-8 Torr were used for all the growths presented here, corresponding to a growth rate of 110nm/h. Diluted disilane was supplied via a custom-built gas delivery system. Disilane was diluted by N2 gas. Two different disilane concentrations were used in our studies, including 0.01wt% and 10wt%. The gas delivery system included an APtech 1410T pressure regulator with two Swagelok PGU-50-P300-L-4FSF ultrahigh-purity pressure gauges, a calibrated Bronkhorst MFC , and a customized three-way pneumatic valve, consisted of two normally closed pneumatic valves APtech AP3540s and AP4540s. Qdisilane was controlled by MFC. The flow rate range for 0.01wt% and 10 wt% disilane was 0-0.705 sccm and 0-0.588 sccm, respectively. In the idle state, the disilane valve was closed, the output pressure was set to zero by the pressure regulator, and the three-way valve was set to “off-state”. A schematic of our hybrid MBE system is shown in figure 1.1 (Ge cell is not shown to save space).

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Annual Report

Investigation of electron transport in β-$(Al,Ga)_2O_3$ thin films and heterostructures under applied high pressures

PI: Elaheh Ahmadi

Award #: FA9550-20-1-0045

Here is a list of tasks performed during this reporting period:

- Demonstration of uniform and controlled Si doping of Ga_2O_3 using diluted disilane
- Enhancement of growth rate using sub-oxide Ga source
- Investigation of electron transport in silicon-doped Ga_2O_3

1. Si doping of β-Ga_2O_3 using diluted disilane

Experimental

All samples were grown in a RIBER 32 hybrid MBE system equipped with conventional Ga and Ge thermal effusion cells. The oxygen source consisted of ultra-high purified oxygen (>99.999%) and was activated by the RIBER RF-O 50/63 oxygen RF plasma source. A plasma power and oxygen flow rate of 410W and 2sccm, respectively, and a Ga flux of 10^{-8} Torr were used for all the growths presented here, corresponding to a growth rate of 110nm/h. Diluted disilane was supplied via a custom-built gas delivery system. Disilane was diluted by N_2 gas. Two different disilane concentrations were used in our studies, including 0.01wt% and 10wt%. The gas delivery system included an APtech 1410T pressure regulator with two Swagelok PGU-50-P300-L-4FSF ultrahigh-purity pressure gauges, a calibrated Bronkhorst MFC , and a customized three-way pneumatic valve, consisted of two normally closed pneumatic valves APtech AP3540s and AP4540s. $Q_{disilane}$ was controlled by MFC. The flow rate range for 0.01wt% and 10 wt% disilane was 0-0.705 sccm and 0-0.588 sccm, respectively. In the idle state, the disilane valve was closed, the output pressure was set to zero by the pressure regulator, and the three-way valve was set to "off-state". A schematic of our hybrid MBE system is shown in **figure 1.1** (Ge cell is not shown to save space).

Fe-doped, semi-insulating, bulk β-Ga_2O_3 (010) substrates were used in our studies. 500 nm Ti layer was deposited on the backside of the substrates for better heat transfer as well as better adhesion to the silicon wafer via In-bonding. The substrates were first diced into 5×5 mm^2 or 5×10 mm^2 pieces and after solvent-cleaning were Indium-bonded to 3-inch Si wafers before being transferred into the growth chamber. The growth was initiated with 30 minutes of oxygen polishing (oxygen flow rate and RF power of 1 sccm and 350 W, respectively) followed by 30 minutes of Ga etching (Ga flux at 1×10^{-8} Torr) conducted at 800 °C to remove the impurities on the substrate surface. A 220 nm unintentionally doped (UID) buffer layer was first grown to separate the Si-doped layer from the substrate interface. It has been shown that Fe from the substrate tends to incorporate into the β-Ga_2O_3 due to the surface riding effect. Fe is a donor trap and compensates free electrons leading to poor electron mobility. Before growing the Si-doped layer, the three-way pneumatic valve in the gas delivery system was set to "off-state" for 15min, to pump out the excess disilane that existed in the gas lines. Then the three-way valve was turned to "on-state", and the disilane was delivered directly to the MBE growth chamber through a low-temperature gas injector cell at room temperature.

The sample surface morphology and surface roughness were studied by Atomic force microscopy (AFM). Secondary ion mass spectrometry (SIMS) was utilized to measure the Si concentration and uniformity as well as unintentional incorporation of impurities such as hydrogen, iron, and carbon. Electron density and mobility of grown samples were determined by Van Der Pauw Hall measurements. For this purpose, indium contacts were made at four corners of each sample.

Results and discussion

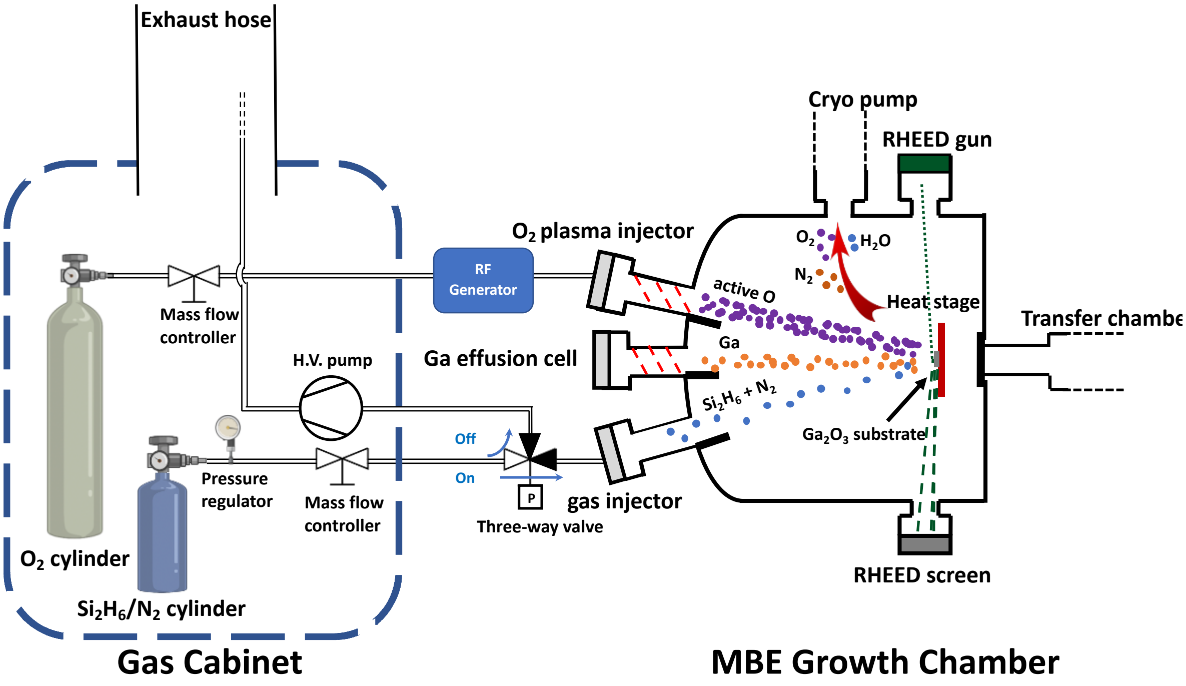

Figure 1.1. Schematic of the hybrid MBE system (Ge cell is omitted). The diluted disilane cylinders is placed in a gas cabinet equipped with gas detectors for saftey. A pressure regulator is directly connected to the disilane cylinder. $Q_{disilane}$ is controlled by an MFC. The disilane gas line is controlled by a normally off three-way pneumatic valve prior to the gas injector. At off-state, the disilane flow is directed to the exhaust line, and the excess disilane existing in the gas line is pumped out by the high vacuum pumping system. During the growth, the three-way valve is set into on-state and the diluted disilane is delivered into the growth chamber.

A SIMS stack was first grown using a 0.01wt% diluted disilane source. This SIMS stack consisted of 220 nm-thick Si-doped β-Ga_2O_3 layers separated by 110 nm-thick UID β-Ga_2O_3 layers. The growth temperature and disilane flow rate were varied for each layer to investigate the dependence of silicon concentration on T_c and $Q_{disilane}$. As shown in **figure 1.2a**, only a slight change in Si concentration, ranging from ~3×10^{16} cm^{-3} to ~6×10^{16} cm^{-3}, was observed when $Q_{disilane}$ was varied from 0.176 sccm to 0.705sccm. To obtain higher Si doping concentrations with enhanced controllability, the gas source was later switched to 10wt% diluted disilane for the rest of the samples discussed in this work. **Figure 1.2b** shows H and Fe profiles. The H incorporation was measured to be ~1×10^{17} cm^{-3} throughout the MBE-grown film and the substrate and did not seem to have any correlation with the disilane flow rate. A Fe tail of ~200 nm can be observed from the SIMS profile which is expected and is due to the Fe incorporated from the substrate as explained earlier. Beyond the initial 200 nm-thick layer, the Fe concentration was at the detecting limit. The sudden jump of H, Si, and Fe at the interface is probably due to substrate surface contamination prior to the growth.

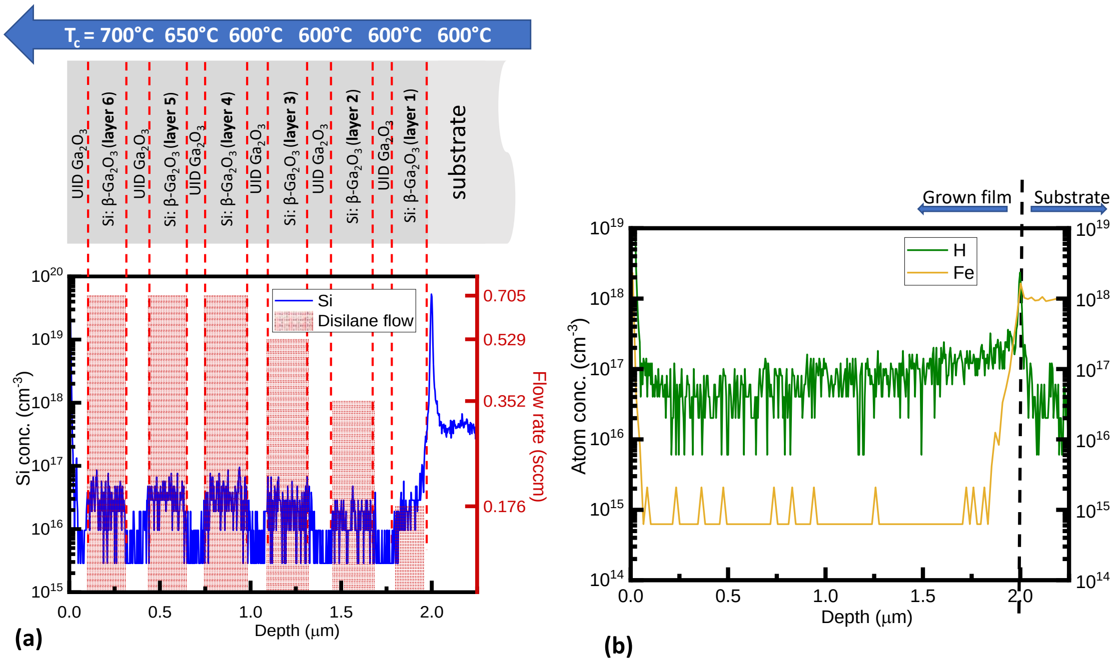

Figure 1.2. (a) schematic of the epi-layer grown for SIMS (b) Si profile in β-Ga_2O_3 doped with 0.01wt% diluted disilane at different T_c (600°C, 650°C, and 700°C) and $Q_{disilane}$ (from 0.176sccm to 0.705sccm). **(b)** H profile and Fe profile measured on the same sample by SIMS.

To further increase silicon incorporation, 10wt% diluted disilane was used. A SIMS stack was first grown at T_c = 525°C, in which Si-doped layers were separated by 110 nm-thick UID Ga_2O_3 films. Si and H profiles are shown in **figure 1.3**. The Si concentration increased from 6×10^{17} cm^{-3} to 2×10^{19} cm^{-3} as $Q_{disilane}$ increased from 0.024sccm to 0.588sccm. Si profile revealed sharp interfaces with a uniform doping plateau along the growth. The H concentration remained constant throughout the MBE-grown film and similar to H concentration in the substrate (~1×10^{17} cm^{-3}), indicating that using disilane as the silicon source did not affect unintentional incorporation of H.

The impact of growth temperature on silicon incorporation was also investigated. **Figure 1.4** shows the Si concentrations of samples grown at different temperatures obtained by SIMS. A $Q_{disilane}$ of 0.588 sccm was used for all four samples shown in this figure. SIMS revealed a relatively sharp and uniform doping profile within the 220 nm-thick silicon doped layer for the growth temperatures ranging from 525°C to 700°C. While a similar silicon concentration of ~1.8×10^{19} cm^{-3} was measured on samples grown at T_c = 600°C, 650°C, and 700°C, the Si incorporation slightly increased to 2.3×10^{19} cm^{-3}. on sample grown at 525°C. It is worth mentioning that the growth rate remained the same (110nm/h) for growth temperatures ranging from 525°C to 700°C.

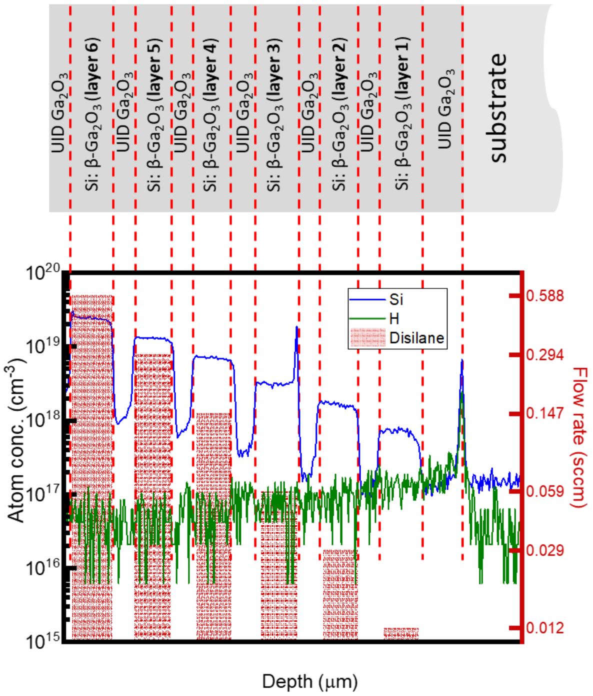

Figure 1.3. Si doping profile of Ga_2O_3 film grown at T_c = 525°C varying $Q_{disilane}$ measured by SIMS . The $Q_{disilane}$ is indicated by the red dashed-dot rectangles for each Si doped layer. $6\times10^{17}cm^{-3}$ to 2×10^{19} cm^{-3} Si concentrations in β-Ga_2O_3 layers were obtained by changing the flow rate from 0.012 sccm to 0.588 sccm.

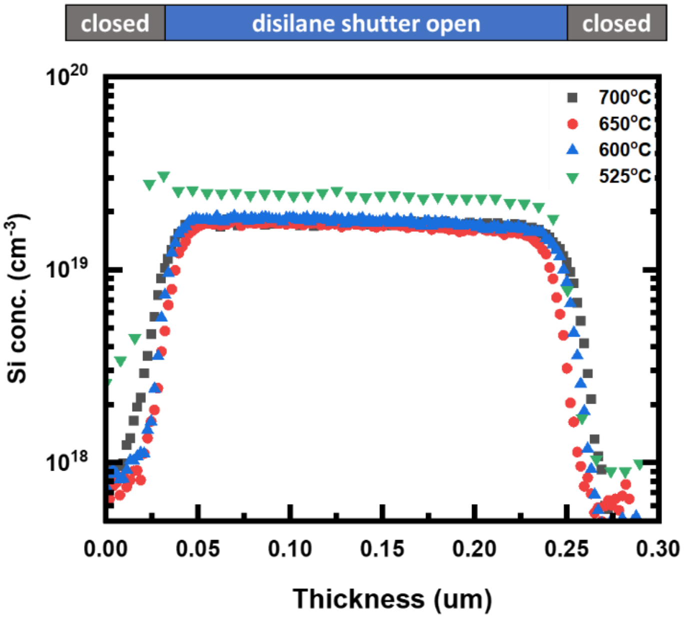

Figure 1.4. Si doping profile of Ga_2O_3 grown at different temperatures, measured by SIMS. Disilane shutter was opened for 2 hours with a flow rate of 0.588 sccm.

In the next step, a series of samples with the epi-structure shown in **figure 5** were grown at different temperatures ranging from 500°C to 700°C, using 10 wt% diluted disilane with $Q_{disilane}$ of 0.029 sccm. A terrace-like surface morphology was observed on all samples as shown in **figure 1.6**. These terraces propagated along β-Ga_2O_3 [100] direction, which is consistent with the previous reports. While the samples grown at T_c = 500°C and T_c = 525°C showed sub-nm surface root mean square (rms) roughness, the surface rms roughness increased significantly as the growth temperature increased due to step bunching. Moreover, the length of traces increased by increasing the growth temperature. This behavior has been previously observed in other step-bunching growth regimes such as MBE-grown Si (111) and MOCVD-grown GaAs (001).

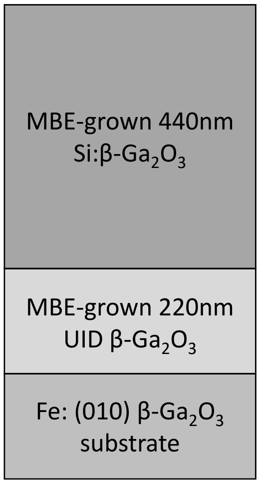

Figure 1.5. Schematic of the epi-structure grown to evaluate electron transport in Si-doped Ga_2O_3 films.

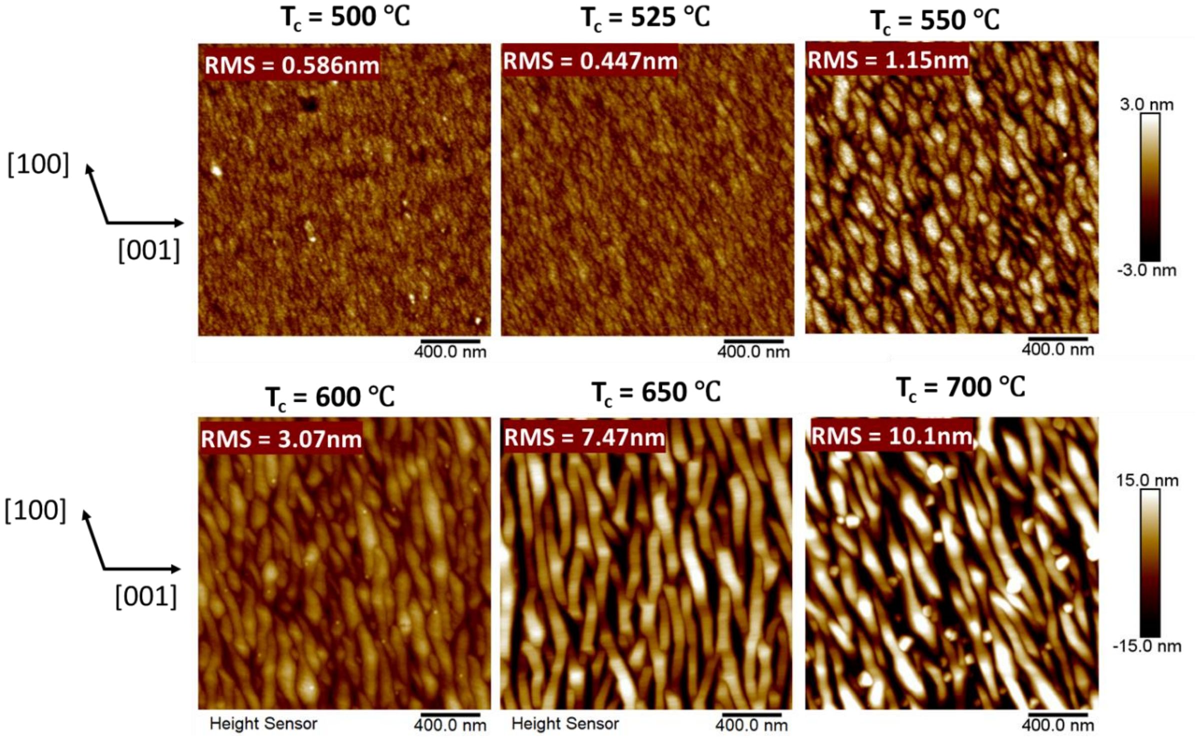

Figure 1.6. 2×2 μm^{2}AFM images of Si doped β-Ga_2O_3 grown with $Q_{disilane}$ = 0.029 sccm at different T_c. RMS surface roughness is indicated. The height profile indication for T_c = 500°C, 525°C, and 550°C is in a range of ±3.0nm, for T_c = 600°C, 650°C and 700°C is ±15.0nm.

In order to evaluate the room temperature electron concentration and mobility, Hall measurements were performed on theses samples (grown at $T_c < 650$°C). Due to poor structural quality and high surface roughness, the samples grown at $T_c \geq 650$°C were highly resistive and difficult to make good ohmic contacts to. Therefore, only samples grown at T_c = 525°C - 600°C were characterized by Hall measurements. The electron mobility and density measured on these samples are reported in **table 1** and illustrated in **figure 1.7**. Carrier concentrations vs. $Q_{disilane}$ at different T_c are shown in **figure 1.7a**. For all growth temperatures, carrier concentration increased as $Q_{disilane}$ increased. For the same $Q_{disilane}$, similar carrier concentrations were measured on samples grown at T_c = 550°C and 600°C. However, samples grown at 525°C had slightly higher carrier concentration, which is consistent with higher silicon incorporation as observed in the SIMS profile **(figure 1.5)**. It is worth noting that the electron concentration in all samples is 20% to 40% lower than the Si concentration. Knowing that Si is a shallow donor in β-Ga_2O_3 (E_d = 16 - 50 meV), a lower carrier concentration than Si concentration suggests the existence of compensating centers in the film, which could be due to point defects formed by either high plasma power or background impurities that lead to deep acceptor levels in β-Ga_2O_3. The carrier mobilities are shown in **figure 1.7b**. The electron mobility reduced as electron density increased for samples grown at T_c = 525°C and T_c = 550°C. This is expected and is due to more ionized impurity scattering in samples with higher doping concentrations. In contrast, for samples grown at 600°C, the electron mobility did not follow a monotonic trend with increase in electron concentration. This non-monotonic behavior could be explained by defect-related compensating centers generated because of high growth temperature. The electron mobility on samples with lower electron density is limited by scattering from such compensating centers. Therefore, higher electron density can help in screening these scattering centers and leads to enhanced electron mobility. However, as doping concentration increases further, the mobility becomes limited by the ionized impurity scattering and therefore reduces as doping concentration increases. **Figure 1.8** summarizes the mobility vs. electron concentration reported for Si, Ge and Sn dopants of β-Ga_2O_3 (010) by different growth techniques. The black bold squares represent our data points. The sample grown at T_c = 525°C and $Q_{disilane}$ = 0.012 sccm showed a record room temperature mobility of 135cm^2/Vs at a carrier density of 3.4×10^{17}cm^{-3}.

Table 1. Room temperature carrier concentration and mobility vs. T_c and $Q_{disilane}$.

	T_c = 525°C		**T_c = 550°C**		**T_c = 600°C**	
$Q_{disilane}$ (sccm)	Carrier conc. (cm^{-3})	Mobility (cm^2/Vs)	Carrier conc. (cm^{-3})	Mobility (cm^2/Vs)	Carrier conc. (cm^{-3})	Mobility (cm^2/Vs)
0.012	3.4×10^{17}	135	2.1×10^{17}	100	2.0×10^{17}	20
0.029	9.6×10^{17}	88	5.8×10^{17}	65	4.5×10^{17}	33
0.059	1.7×10^{18}	83	1.2×10^{18}	62	1.1×10^{18}	79
0.147	4.3×10^{18}	45	2.5×10^{18}	60	4.5×10^{18}	38
0.294	8.1×10^{18}	30	7.7×10^{18}	55	7.0×10^{18}	48
0.588	1.8×10^{19}	20	1.4×10^{19}	34	1.1×10^{19}	66

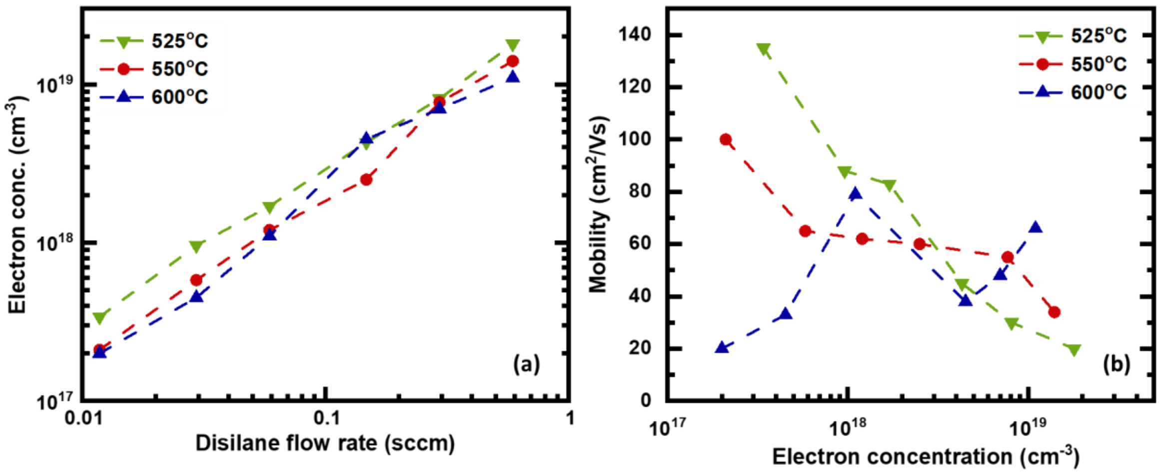

Figure 1.7. (a) Hall measurement carrier concentration vs. different disilane flow rates for samples grown at T_c = 525°C, 550°C, and 600°C. **(b)** Hall mobility vs. carrier concentration for samples grown at T_c = 525°C, 550°C, and 600°C.

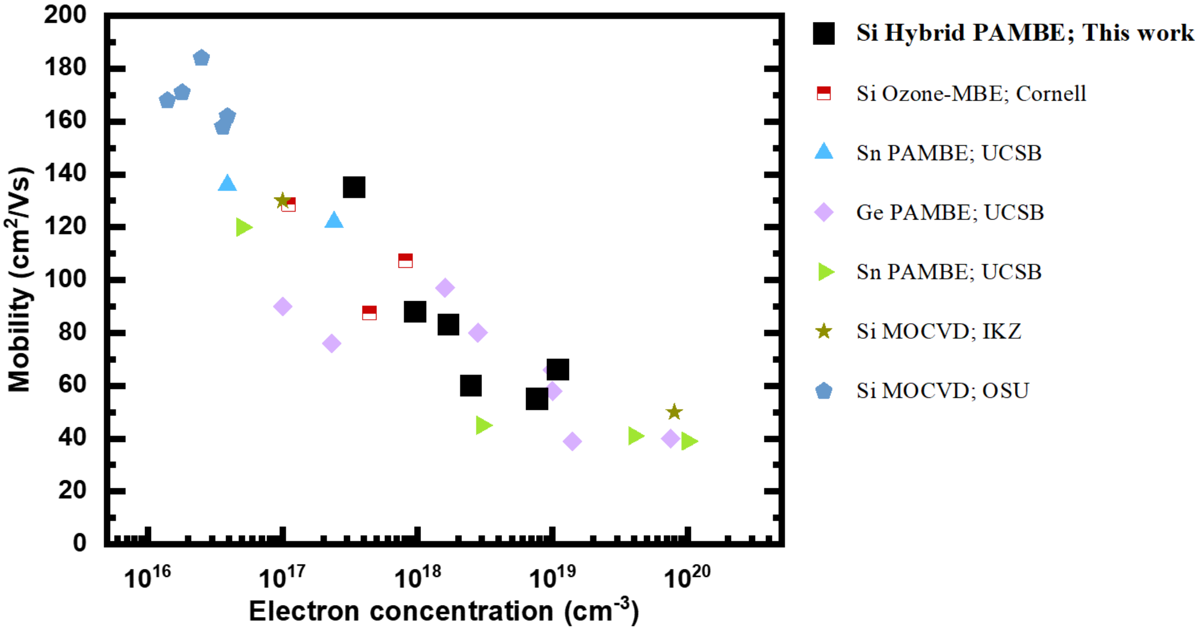

Figure 1.8. The summary of reported mobility vs. electron concentration for different dopants of β-Ga_2O_3 (010) by various growth techniques.

Sub-oxide Ga source to enhance growth rate

Traditionally, Ga_2O_3 is grown by using Ga elements and active O species in the MBE system. The reaction is as follow:

$$2Ga(g) + 3O_2(g) = Ga_2O_3(s) \tag{1}$$

The growth rate of Ga_2O_3 in MBE system is quite limited due to the desorption of Ga_2O, which forms when increasing the Ga flux and not enough active oxygen is available to oxidize.

$$Ga(g) + Ga_2O_3(s) = 3GaO(g) \tag{2}$$

Recently, it is reported in the ozone MBE system that the low growth rate of Ga_2O_3 can be improve by directly grow from Ga_2O and oxygen.

$$4Ga + Ga_2O_3(s) = 3Ga_2O(s) \tag{3}$$

The growth rate of Ga_2O_3 in ozone MBE system was increased by one order of magnitude up to beyond 1μm/h (**figure 1**).

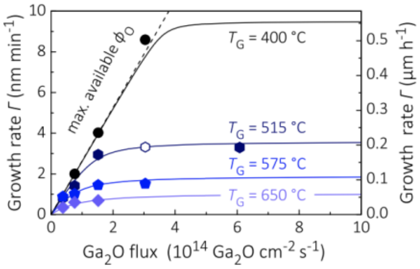

Figure 2.1. Reported growth rate of Ga_2O_3 in ozone MBE system using Ga_2O suboxide as sources at different growth temperatures.

We have started to study suboxide growth of Ga_2O_3 in PAMBE system (s-PAMBE). **Figure 2** shows a SIMS profile of a stack grown using suboxide Ga source in hybrid PAMBE system. The sample was grown at 550 °C with 5x10^7Torr Ga_2O flux. The oxygen plasma is 410W, 2sccm. By controlling the disilane flow rate ($Q_{disilane}$), doping concentrations ranging from several 10^{16}cm^{-3} to 10^{19}cm^{-3} has been achieved. The growth rate using suboxide source was enhanced to ~200nm/h, which is doubled than that of using traditional Ga source. The Si doping concentration is further reduced at same $Q_{disilane}$ with using suboxide compared with that in layers grown using elemental Ga. **Figure 3** shows the Ga_2O_3 surface morphology under AFM microscopy with different Ga_2O fluxes at 600°C. They all show a typical Ga_2O_3 terrace surface morphology with a root mean square surface roughness ~1nm. Steaky RHEED patterns were observed during the growth of all the samples, indicating atomically smooth surface.

The electron transport was also studied using hall measurements. **Figure 4** shows a Si doping series grown at 550°C. The carrier concentration could be tuned from 5x10^{17}cm^{-3} to 1x10^{19}cm^{-3} with different $Q_{disilane}$. However, the electron mobilities under low carrier concentrations are relatively low. Further studies are necessary to understand the reasons for low electron mobility and . **Figure 5** shows some preliminary data of low carrier concentrations vs. mobility under different growth parameters. The testing structure was grown with 200nm UID separation layer with Ga source, 300nm Si doping layer with suboxide source, and another 20nm highly doped layer with Ga source. Ohmic contacts were made by 20nm Ti/200nm Au metal deposition at the corner of the surface before etching down the 20nm highly doped layer. Hall measurements show that the electron density in the range of 10^{16}cm^{-3} can be achieved by using suboxide growth of Ga_2O_3. The mobilities are relatively low compared with literature data and further work on the improvement is necessary.

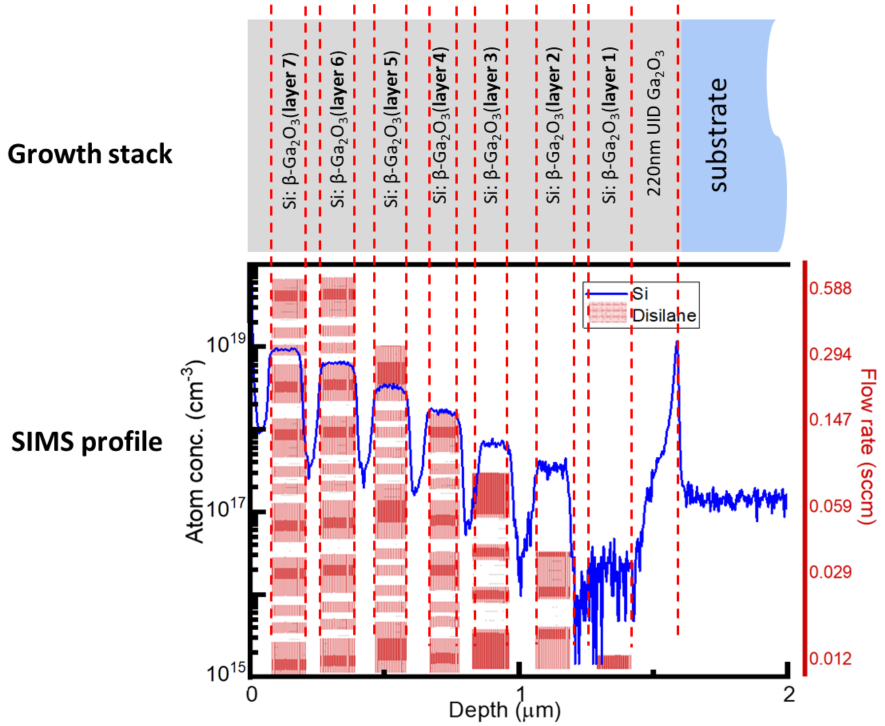

Figure 2.2. SIMS stack of Si doped Ga$_2$O$_3$ grown by s-PAMBE. 10% dilluted disilane was used as Si source for doping. The Si doping can be well-controlled by Q$_{disilane}$. Each doping layer was grown for 40min.

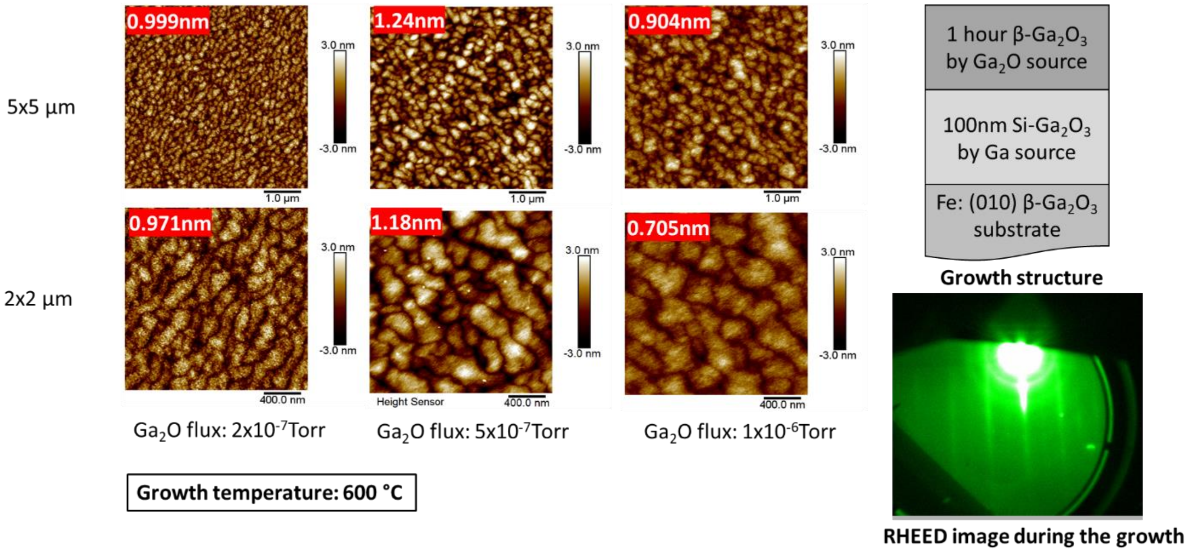

Figure 2.3. AFM images of Ga$_2$O$_3$ grown by suboxide at 600°C. Steaky RHEED patterns were observed.

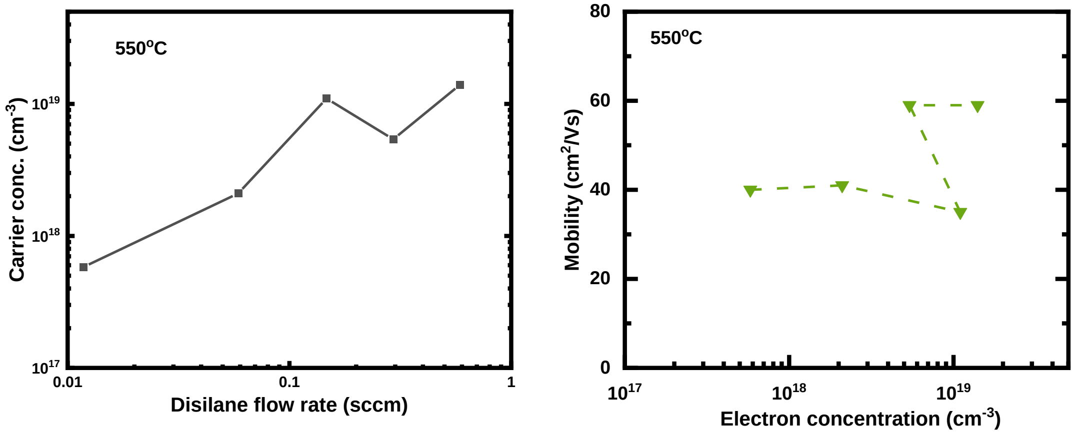

Figure 2.4. Hall measurements of suboxide growth Si-doped Ga_2O_3 samples grown at 550°C.

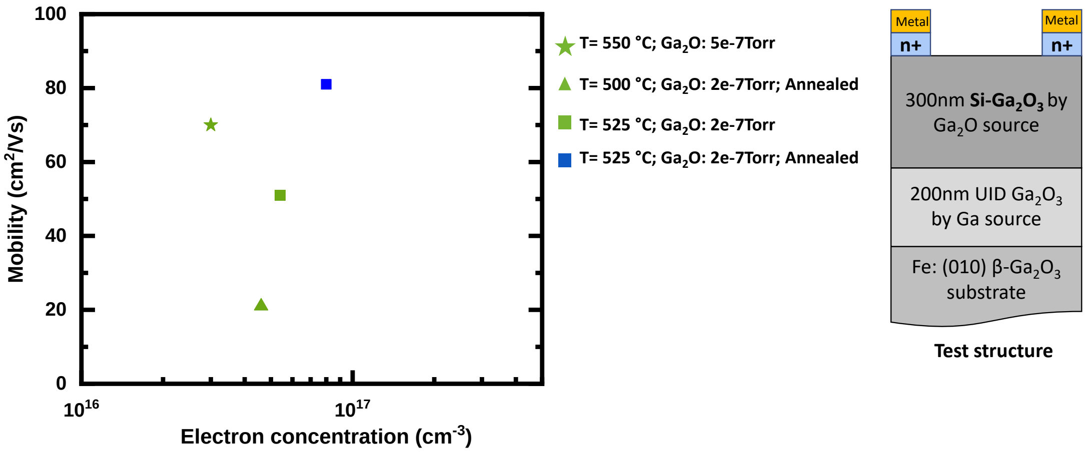

Figure 2.5. Electron concentrations vs. mobilities of lightly Si-doped Ga_2O_3 samples.