Surface Treatment for Stepped Vicinal (100) β-Ga_2O_3 Substrates for Homoepitaxial Growth by S-MBE

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Abstract

β-Ga_2O_3 is an ultrawide bandgap (UWBG) semiconductor of distinct interest for its high breakdown electric fields and its unique ability among UWBG semiconductors to be grown using the Czochralski method, allowing for the bulk synthesis of doped substrates.[1] Homoepitaxial film growth of β-Ga_2O_3 on the (100) plane has been historically limited by the formation of twin boundaries, which limit the electron mobilities of devices. This work investigates the ideal cleaning, etching, and annealing of various vicinal (100) β-Ga_2O_3 substrates to obtain a smooth step-terrace surface with RMS roughness values of <600 pm for the reduction of twin boundaries in films grown by suboxide-molecular beam epitaxy.

Introduction

Quality of homoepitaxial thin-film β-Ga_2O_3 growth along the (100) plane by various methods has been demonstrated to be limited by the formation of incoherent twin boundaries,[2] an issue theoretically suppressed by the usage of highly vicinal stepped substrates.[3] Commercially available semi-insulating vicinal substrates of β-Ga_2O_3 are limited by surface contaminants and a lack of step-terrace structures, so this study develops and refines methods of cleaning, wet etching, and annealing for the preparation of high-quality stepped substrates for epitaxial growth.

Methods

Semi-insulating vicinal gallium oxide substrates doped with magnesium and iron were purchased from CrysTec and Novel Crystal Technology (NCT), respectively. CrysTec Mg-doped substrates of miscut angles 2°, 3°, 4°, and 6°, and NCT Fe-doped substrates of miscut angles 1.8°, 3.5°, and 5.9°, were used in this study. All substrates are miscut towards the $(00\bar{1})$ direction to create step facets that are not susceptible to twinning.[3]

Out of box substrates were first sonicated in consecutive solutions of acetone, isopropanol, and deionized water for 30 minutes each, followed by a spin-rinse in DI water to clean their surfaces of large particles and macroscopic surface contaminants such as dust.

Mg-Ga_2O_3 substrates were etched with hot H_3PO_4 at 140° Celsius,[4] while Fe-Ga_2O_3 substrates were etched with 47% HF at room temperature,[5] as previous reports have demonstrated successful etching of β-Ga_2O_3 at these conditions. An etch time of 15 minutes was determined to fully etch the surface layers without the formation of deep cavities on the surface.

Surface morphologies at each step of the preparation process were studied using atomic force microscopy (AFM) images collected by an Asylum Research Cypher S SPM system operating in AFM tapping mode at a scan rate of 3.91 Hz and using a 10 nm radius tip.

Results

Figure 1 demonstrates the successes of the cleaning (Fig. 1, a-b) and acid etch processes (Fig. 1, c-f) as previously detailed in the removal of most surface contaminants by AFM. The hot phosphoric acid etch, as utilized in the preparation of magnesium-doped substrates, shows the most success in the creation of a smooth etched surface for both magnesium- and iron-doped substrates, although the use of phosphoric acid on an iron doped substrate consistently leads to the development of significant surface particles after O_2 annealing, as seen in (Fig. 1, g-h). This may either stem from the differences in dopant, or a difference in defect structure arising from growth methods (Mg-doped CrysTec: Czochralski, Fe-doped NCT: EFG), though further investigation will be required to understand this mechanism. While the HF etch demonstrates general increases in the surface roughnesses, we believe that it is still an essential step for the removal of contaminants that cannot be removed during the O_2 anneal.

The ideal annealing length, t, for vicinal β-Ga_2O_3 substrates can be fit to experimental data using the geometry of the terraces. Assuming that the diffusion length of surface atoms, which is proportional to $\sqrt{t}$, should be proportional to the terrace width and given a consistent step height of approximately 0.59 nm,[3] $t \propto \cot(\theta)^2$.

Inital conditions for the O_2 anneal were derived from R. Schewski et al.,[3] and the temperature of 900 °C was verified while the lengths of the anneals were refined for our miscut angles. Short or cooler anneals demonstrated limited step formation, while long or hotter anneals demonstrated breaking of steps along their length as a result of the cavities present on the surface after the etch. The success of anneals across the range of miscuts from 1-6° follow the trends as expected by the proposed model, and a 3 hour, 900 °C successful anneal on a 2° Mg-doped substrate was utilized to fit the model:

$$t(hr) = 0.00366{*}\cot(\theta)^2$$

Successful cleaning, wet etching, and annealing of substrates of miscut angles 1.8° through 5.9° are demonstrated in Figure 2, alongside shaded AFM images which highlight the shapes of step formation. All of these sub-

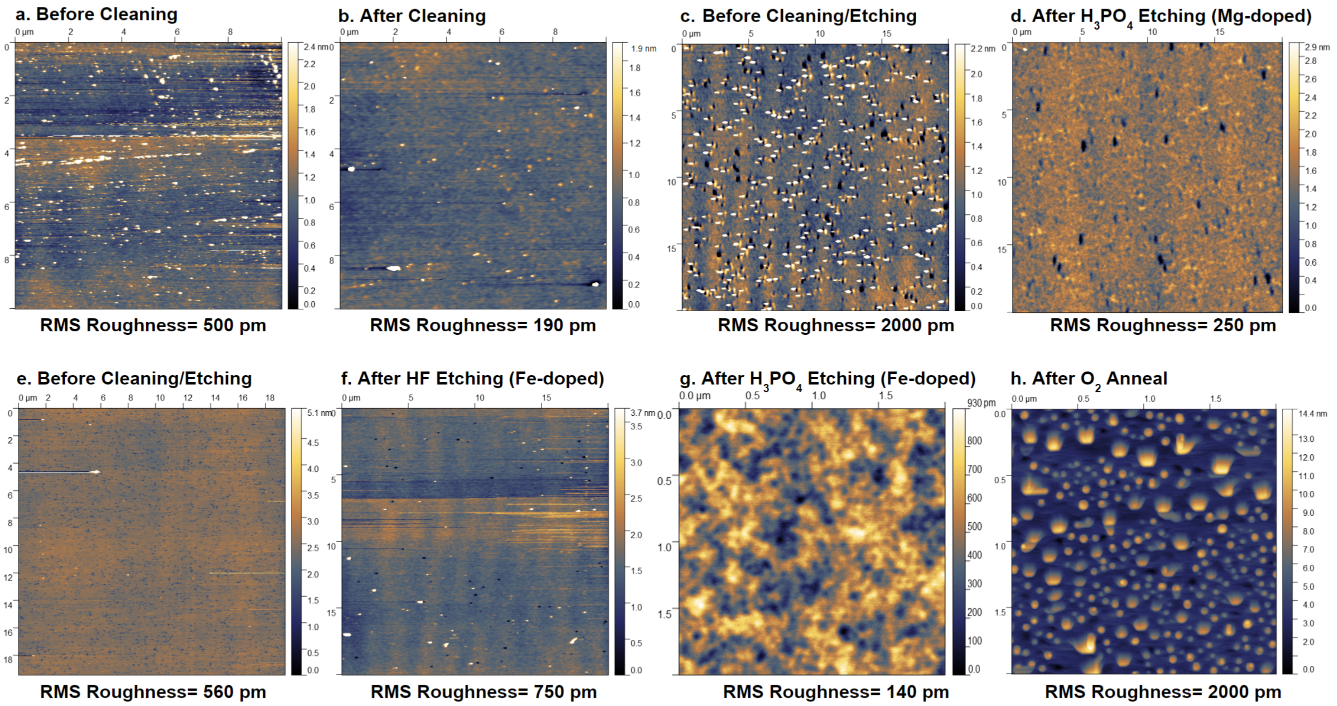
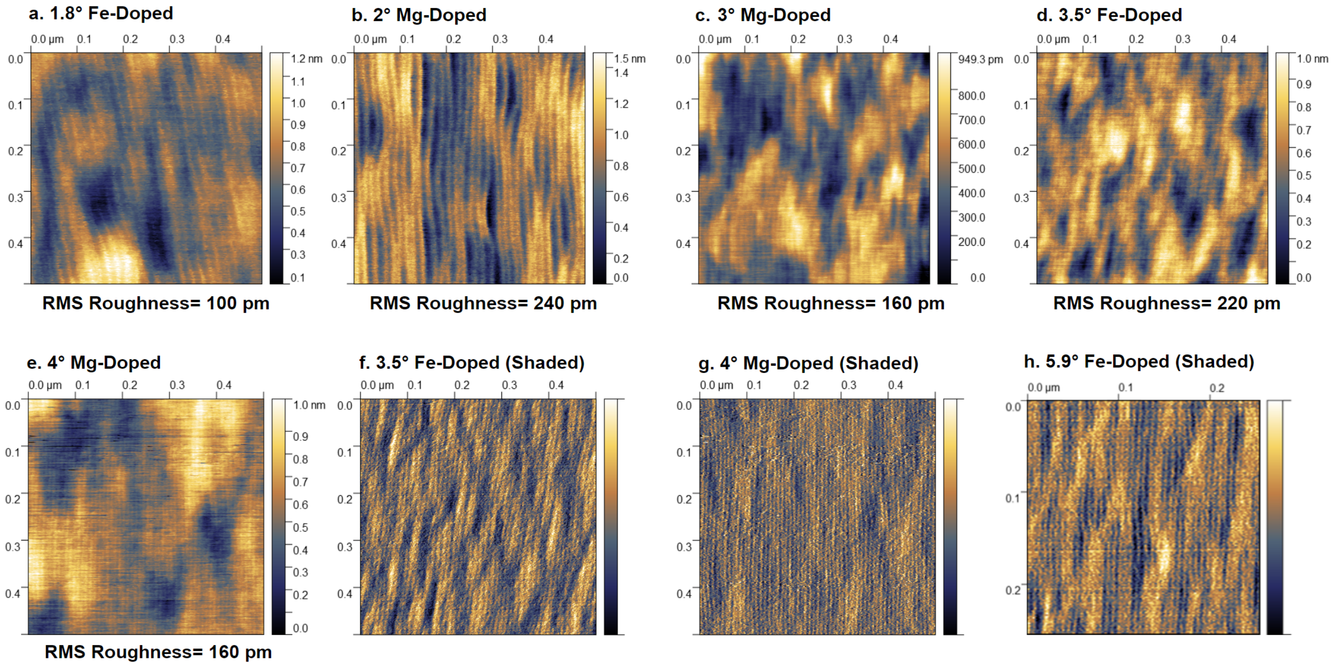


Figure 1: Cleaning and etching process from typical out-of-box substrate (a). (a-b) demonstrate the acetone/isopropanol/water cleaning process, (c-d) demonstrate the cleaning and phosphoric acid etch on a magnesium-doped substrate, (e-f) demonstrate the cleaning and hydrofluoric acid etch on an iron-doped substrate, and (g-h) demonstrate the failure of phosphoric acid etching on an iron-doped substrate after anneal.


a. 1.8° Fe-Doped

RMS Roughness= 100 pm

b. 2° Mg-Doped

RMS Roughness= 240 pm

c. 3° Mg-Doped

RMS Roughness= 160 pm

d. 3.5° Fe-Doped

RMS Roughness= 220 pm

e. 4° Mg-Doped

RMS Roughness= 160 pm

f. 3.5° Fe-Doped (Shaded)

g. 4° Mg-Doped (Shaded)

h. 5.9° Fe-Doped (Shaded)

Figure 2: Annealing process following the derived model. Substrates were annealed at 900 °C for (a) 3.7 hr, (b) 3 hr, (c) 1.3 hr, (d/f) 1 hr, (e/g) 0.75 hr, (h) 0.34 hr. Shaded AFM scans (f-h) allow for viewing of fine step features, though remove numerical height accuracy of overall surface. Steps were only visible on 5.9° substrates using shading, and future work will improve imaging of high vicinalities where the steps are smaller than the AFM tips in use.

strates demonstrate successful step formation with step heights of 0.59 ± 0.05 nm and RMS roughness values <600 pm on 20 μm x 20 μm AFM scans.

Conclusions and Future Work

We demonstrate the successful cleaning, wet etching, and annealing of (100) vicinal semi-insulating β-Ga_2O_3 substrates doped with magnesium or iron. The use of isopropanol, acetone, and DI water in cleaning and the use of wet etching in H_3PO_4 (CrysTec Mg-doped substrates) or HF (NCT Fe-doped substrates) demonstrate the successful removal of surface contaminants with low surface roughness. We demonstrate a successful recipe for 900 °C anneals with lengths dependent on miscut angle, forming straight, consistent-height steps on a range of miscut angles from 1.8° through 5.9°, paving the way for future high-quality epitaxial growth.

Future studies will apply these high-quality substrates in suboxide molecular beam epitaxy for the growth of Si-Ga_2O_3 at silicon doping concentrations relevant for devices. Using these films, we will investigate the suppression of twin boundary formation and the resulting mobilities of films as a function of miscut angle.

Acknowledgements

I would like to acknowledge the help of Kathy Azizie and Darrell G. Schlom for their support throughout this project, and Hari Nair for his collaborations on the theory in this work. This work was supported by the National Science Foundation (REU Site: Summer Research Program at PARADIM) under Cooperative Agreement No. DMR-2150446, and by the AFOSR/AFRL ACCESS Center of Excellence under Award No. FA9550-18-10529.

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