# “High Throughput” Exploration of Oxide MBE Growth Space through Cyclical *in situ* Growth and Etching

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Beta phase gallium oxide ($\beta$-$Ga_2O_3$) is an emerging ultra-wide bandgap semiconductor that has attracted attention for its potential to outperform existing materials operating at high breakdown voltages and high temperature. Alloying of In and Al in $\beta$-$Ga_2O_3$ provides the ability to individually engineer the bandgap and lattice parameters of the material, providing a useful toolbox for heterostructure engineering. However, the tendency of $(Al,In,Ga)_2O_3$ alloys to form competing phases, along with the complex suboxide chemistry of Ga and In, results in a growth window that is difficult to map and an alloy which is difficult to control.

We report on a high-throughput molecular beam epitaxy (MBE) technique to screen the growth conditions for the ternary alloy $(In_yGa_{1-y})_2O_3$, and the application of these findings to the successful synthesis of monoclinic $(Al_xGa_{1-x-y}In_y)_2O_3$. By leveraging the sub-oxide chemistry of $Ga_2O_3$ and *in-situ* monitoring by reflection high-energy electron diffraction (RHEED), a cyclical growth and etch-back method is developed to rapidly characterize the $(In_yGa_{1-y})_2O_3$ growth space. This cyclical method provides approximately 10x increase in experimental throughput and 46x improvement in $Ga_2O_3$ substrate utilization. Growth conditions for monoclinic $(In_yGa_{1-y})_2O_3$ are identified and targeted growths are characterized *ex-situ* to confirm improved In incorporation. These conditions are then used to grow quaternary $(Al_xGa_{1-x-y}In_y)_2O_3$ with Al cation composition $x$ ranging from 1% – 24% and In cation composition $y$ ranging from 3% to 16%. The structural, chemical and optical properties of the alloys are investigated. An $(Al_{0.17}Ga_{0.76}In_{0.07})_2O_3$ alloy lattice-matched to $Ga_2O_3$ is examined by high resolution microscopy, highlighting the correlation between surface facets and composition. Such lattice-matched material can be grown arbitrarily thick without elastic strain and relaxation, making it suitable for high voltage diodes, transistor barriers, and epitaxial dielectrics.

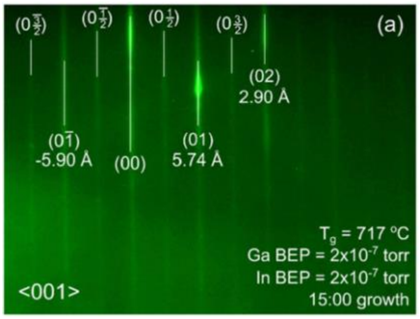

Figure 1: RHEED image typical of In-catalyzed $Ga_2O_3$ growth

[1] S. Schaefer, J. Mater. Chem. A, **12**, 5508(2024).

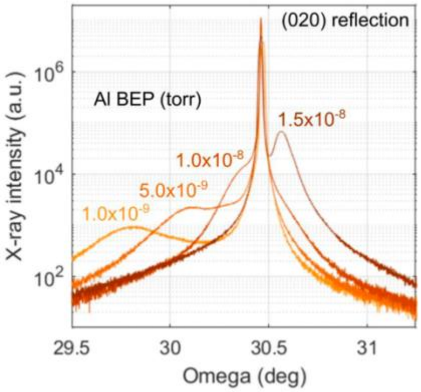

Figure 2: X-ray diffraction of $(Al,In,Ga)_2O_3$ alloys grown at various Al flux values.

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# Suplementary Pages (Optional)

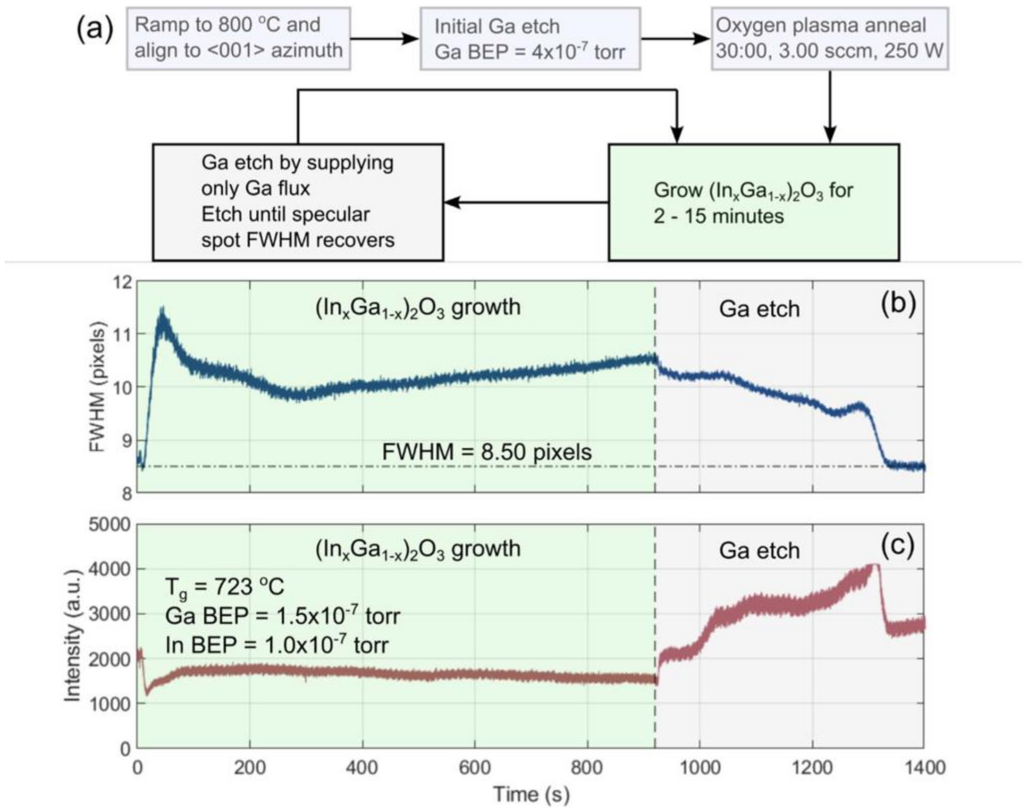

Figure S1: Overview of cyclical growth and etch process. (a) Flow diagram showing process of growth and etching. (b) FWHM of the specular RHEED reflection during the growth and etch-back process. (c) RHEED spot intensity during the growth and etch-back process.

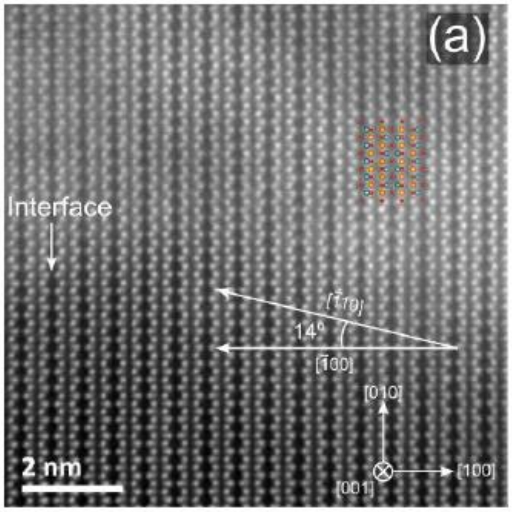

Figure S2: Scanning transmission electron microscopy (STEM) demonstrating beta-phase $(Al_{0.17}Ga_{0.76}In_{0.07})_2O_3$

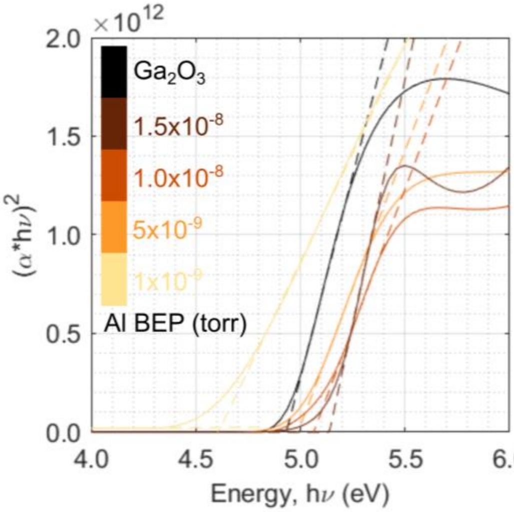

Figure S3: Spectroscopic ellipsometry Tauc analysis of $(Al,In,Ga)_2O_3$ optical absorption onset.