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Atomic Distribution of Al and Phase Transformation in β-$(Al_xGa_{1-x})_2O_3$

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β-Ga_2O_3 has gained tremendous attention as a promising transparent conductive oxide, ultra-wide band material due to its unique advantages including a large band gap energy (~ 4.8eV), high breakdown voltage, and its availability as high quality bulk grown single crystals [1]. However, realizing its full potential as a semiconductor material requires the manipulation and control of its band gap, which could enable novel heterostructure designs for carrier confinement and field effect transistors or deep UV optoelectronic devices. Recent efforts have indicated that the bandgap of β-Ga_2O_3 can be engineered through the incorporation of Al [2], forming β-$(Al_xGa_{1-x})_2O_3$ with a band gap up to 8.8 eV. High crystalline quality β-$(Al_xGa_{1-x})_2O_3$ structures with Al composition up to x=40% have been achieved using metalorganic chemical vapor deposition (MOCVD) [1] and a high mobility two-dimensional electron gas has been demonstrated to form at a β-$(Al_xGa_{1-x})_2O_3$/Ga_2O_3 interface [2]. Ultimately, advancing these alloyed β-Ga_2O_3 epitaxial films requires the precise control of the point defects that critically influence their important properties. And their exact control requires attaining the essential detailed atomic scale structural information, which will provide key information on their formation during growth and their impact on properties.

Here, we present the atomic scale characterization of β-$(Al_xGa_{1-x})_2O_3$ and the observation of phase transformation in $(Al_xGa_{1-x})_2O_3$ thin films. Employing the unprecedented resolution and chemical sensitivity generated by quantitative scanning transmission electron microscopy (STEM) [3], we specified the exact substitutional positions of the alloying-element Al in molecular beam epitaxy (MBE) and MOCVD grown β-$(Al_xGa_{1-x})_2O_3$ films (Fig. 1). Quantitative Al site occupancy results were then correlated with density function theory (DFT) and strain information acquired through electron nanodiffraction to better understand the critical thicknesses of β-$(Al_xGa_{1-x})_2O_3$ films for superlattice structure synthesis. Additionally, quantitative STEM was performed to detail planar defect structures that develop with increasing Al content within β-$(Al_xGa_{1-x})_2O_3$ films (Fig. 1(e) and (f)). These defect structures revealed a configuration similar to the recently discovered divacancy – interstitial complexes found in β-Ga_2O_3 bulk crystals [4]. To provide an accurate identification of the defect structures, the exact position and composition of each atomic column was determined and then utilized by DFT to reveal their electronic behavior. Furthermore, the mixture of β and γ phases in $(Al_xGa_{1-x})_2O_3$ thin films were investigated using STEM imaging and nanodiffraction (Fig. 2). Driven by the strain induced from the configurational relaxation during Al incorporation, the rotations of the β phase and introduction of the defective spinel structure were identified. In this STEM investigation, we obtained critical atomic scale defect information that provides guidance to the growth of β-$(Al_xGa_{1-x})_2O_3$ with controlled properties.
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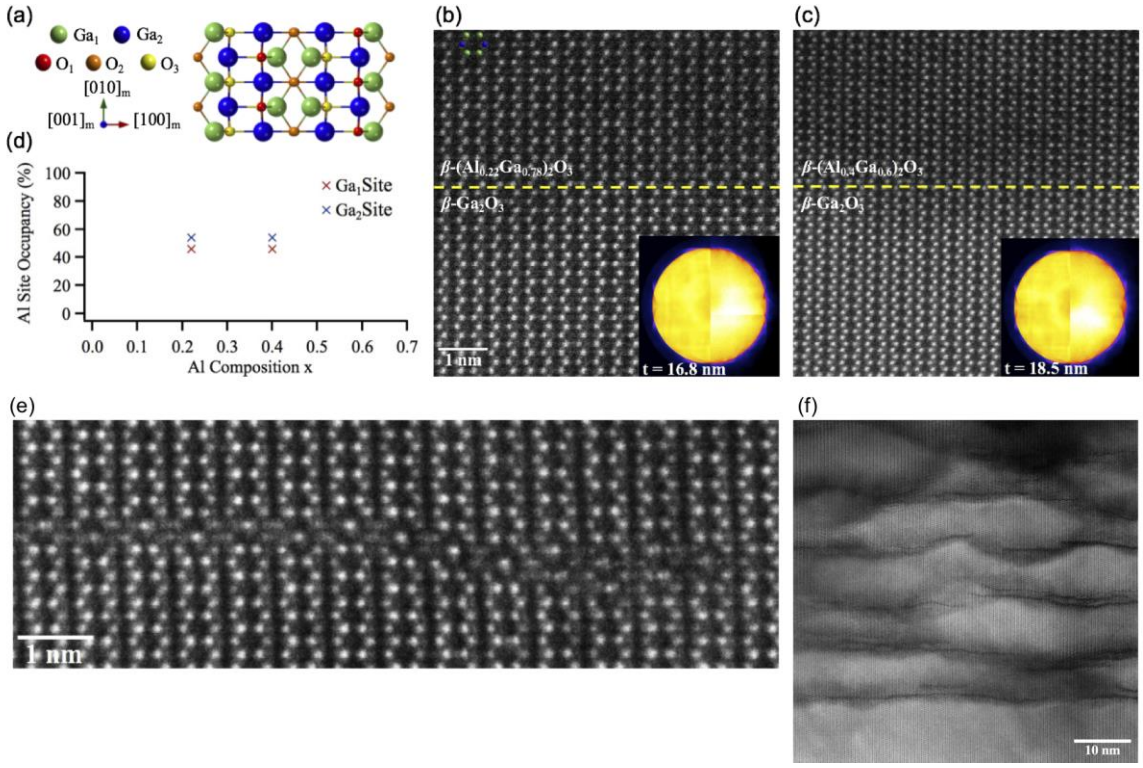

Figure 1. (a) Crystal structure of β-Ga2O3 along the [001]m direction. (b) and (c) HAADF STEM images of (b) β-(Al0.22Ga0.78)2O3 and (c) β-(Al0.40Ga0.60)2O3 films used for the quantitative analysis of Al site occupancy. Inset are the experimental PACBED patterns acquired from the β-Ga2O3 substrate and the corresponding multislice simulated patterns, indicating a 16.8 nm and 18.5 nm thick TEM sample for the β-(Al0.22Ga0.78)2O3 and β-(Al0.40Ga0.60)2O3 films, respectively. (d) Quantitative STEM data showing the Al site occupancy (%) in the Ga1 (red x) and Ga2 (blue x) sites versus the Al composition of the film. The HAADF STEM images reveal planar defects in (e) a β-(Al0.40Ga0.60)2O3 epitaxial film and (f) a β-(Al0.56Ga0.44)2O3 superlattice structure.

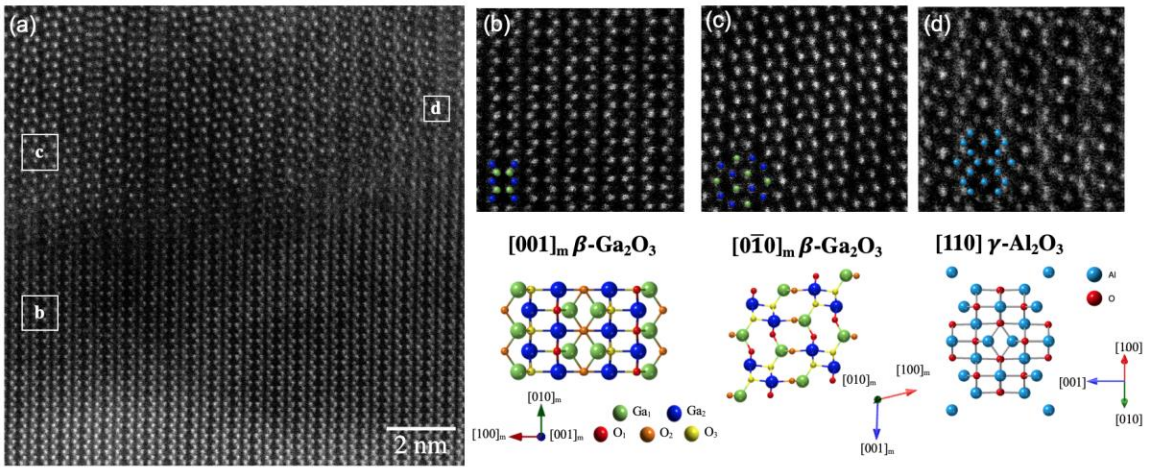

Figure 2. Atomic resolution HAADF STEM images of the (Al0.40Ga0.60)2O3 film grown on β-Ga2O3 substrate. (a) The first 12 nm of the (Al0.40Ga0.60)2O3 film demonstrating three distinct growth structures. (b) (010) β-(Al0.40Ga0.60)2O3 growth with the model orientated along the [001]m imaging direction. (c) (001) β-(Al0.40Ga0.60)2O3 growth displaying a 90° rotation of the (010) β-

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(Al0.40Ga0.60)2O3 and a model oriented along the corresponding [010]m imaging direction. (d) (110) γ-(Al0.36Ga0.64)2O3 phase transformation region with a model of [110] γ-Al2O3.

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