Band Alignment of Atomic Layer Deposited SiO$_2$ and Al$_2$O$_3$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ for x = 0.2-0.65

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(Al$_x$Ga$_{1-x}$)$_2$O$_3$ is attracting attention for use in heterostructure devices grown on Ga$_2$O$_3$ substrates. The band alignments of amorphous, atomic layer deposited Al$_2$O$_3$ and SiO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ for x = 0.2-0.65 have been determined using high resolution X-ray photoelectron spectroscopy. The (Al$_x$Ga$_{1-x}$)$_2$O$_3$ was grown by continuous composition spread Pulsed Laser Deposition (CCS-PLD). The band alignments are type I (nested gap) in all cases, with conduction band offsets ranging from 1.57-0.67 eV for Al$_2$O$_3$ on (Al$_{0.2}$Ga$_{0.8}$)$_2$O$_3$ to (Al$_{0.65}$Ga$_{0.35}$)$_2$O$_3$ and 2.35-1.40 eV for SiO$_2$ on these same compositions. Correspondingly, the valence band offsets are all >1.25 eV for SiO$_2$ and 0.23-0.33eV for Al$_2$O$_3$ over this composition range. While these are the first reports for Al$_2$O$_3$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ over such a wide composition range, our results differ by up to 0.4 eV in conduction band offsets from past studies of SiO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ of a more limited composition range, which themselves have shown variations of up to 0.5eV for conduction band offsets on nominally the same composition. These differences emphasize the influence of experimental conditions in determining band alignments.
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The maximum solubility of Al in β-Ga$_2$O$_3$ is generally reported to be in the range 67–78%.[1–5] When grown by a variety of methods, including pulsed laser deposition and molecular beam epitaxy.[6–14] Methods for calculating the strain in pseudomorphic (Al$_x$Ga$_{1-x}$)$_2$O$_3$ heterostructures on bulk β-Ga$_2$O$_3$ substrates are also available.[11,12] There have been recent demonstrations of high quality (Al$_x$Ga$_{1-x}$)$_2$O$_3$/Ga$_2$O$_3$ heterostructures for field effect transistors with enhanced electron mobility due to formation of two-dimensional electron gases (2DEG) at the heterointerface.[14–21] For example, Zhang et al.[21] reported formation of a 2DEG in a modulation-doped β-(Al$_x$Ga$_{1-x}$)$_2$O$_3$/Ga$_2$O$_3$ structure from Hall measurements, with channel mobility of 143 cm^2/V·s at 300K and 1520 cm^2/V·s at 50 K. Such devices, if optimized, might be useful for RF power device applications. The (Al$_x$Ga$_{1-x}$)$_2$O$_3$ can also be used in solar-blind UV photodetectors.[22,23]

A key requirement in MOS-gate wide bandgap power devices is that the gate dielectric have sufficient band offsets to ensure good carrier confinement at the heterointerface with the semiconductor. A schematic of a prototypical device embodiment is shown in Figure 1, where the dielectric is used to form a MOS gate on the (Al$_x$Ga$_{1-x}$)$_2$O$_3$. The dielectric needs to be thermodynamically stable on the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ and have sufficient band offsets to produce good carrier confinement. There are only a few reports of band alignments for dielectrics on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ at a limited range of Al contents[24,25] or even just a single Al concentration (0.14, typical of heterostructure transistor structures).[26–29] Feng et al.[24] determined the band alignment for atomic layer deposited (ALD) SiO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ with x = 0-0.49. The valence band offsets were in the range 1.5-0.8 eV for this composition range. They also reported band offsets for ALD deposited SiO$_2$ and HfO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ with x = 0-0.53.[25] Even within these studies from the same group, differences of up to 0.5 eV in conduction band offset and 0.3 eV in valence band offset were found for SiO$_2$ on nominally similar Al contents in the (Al$_x$Ga$_{1-x}$)$_2$O$_3$. Such differences are not uncommon in the literature on band offsets on semiconductors and have been ascribed to the effects of dielectric deposition method on surface stoichiometry and defect density, bandgap of the dielectric, and contamination.[30] It is clearly of value to examine the band alignments over a wider range of Al contents, to measure this for Al$_2$O$_3$

because of its compatibility with (Al$_x$Ga$_{1-x}$)$_2$O$_3$ and also to measure the phase purity of the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ used in the experiments.

In this paper, we utilize X-Ray Photoelectron Spectroscopy (XPS) to determine the valence band offsets in SiO$_2$ and Al$_2$O$_3$/ (Al$_x$Ga$_{1-x}$)$_2$O$_3$ heterostructures with x = 0.2−0.65, in which amorphous dielectrics were deposited by ALD onto (Al$_x$Ga$_{1-x}$)$_2$O$_3$ grown by continuous composition spread Pulsed Laser Deposition (CCS-PLD).[1,13] In these films, the monoclinic phase is phase pure up to ∼40-45 at.%, then a mixed phase with γ-(Al$_x$Ga$_{1-x}$)$_2$O$_3$ is present. Above roughly 50–55 at.%, the thin film shows purely the γ-phase. SiO$_2$ is found to have both conduction band and valence band offsets >1.2 eV over the entire composition range examined, while Al$_2$O$_3$ has relatively small valence band offsets (eV) over this composition range.

Experimental

The (Al$_x$Ga$_{1-x}$)$_2$O$_3$ films with x = 0.15-0.70 were grown on (100) MgO substrates by continuous composition spread Pulsed Laser Deposition (CCS-PLD), which relies on the ablation of segmented PLD targets.[13,31] This CCS-PLD method can be used in existing off-set PLD systems without any modification of the hardware. A spatial offset between the substrate center and the centerline of the expanding plasma plume and a synchronized rotation of substrate and semicircular-segmented targets (lateral target segmentation) are used to obtain a lateral continuous composition spread.[1,13] The distribution of the different elements originating from different target segments on the substrate depends on background pressure, target-to-substrate distance, and the offset, as well as the normal thermodynamic conditions for deposition.[31]
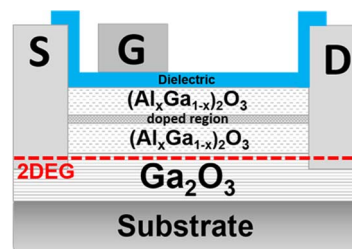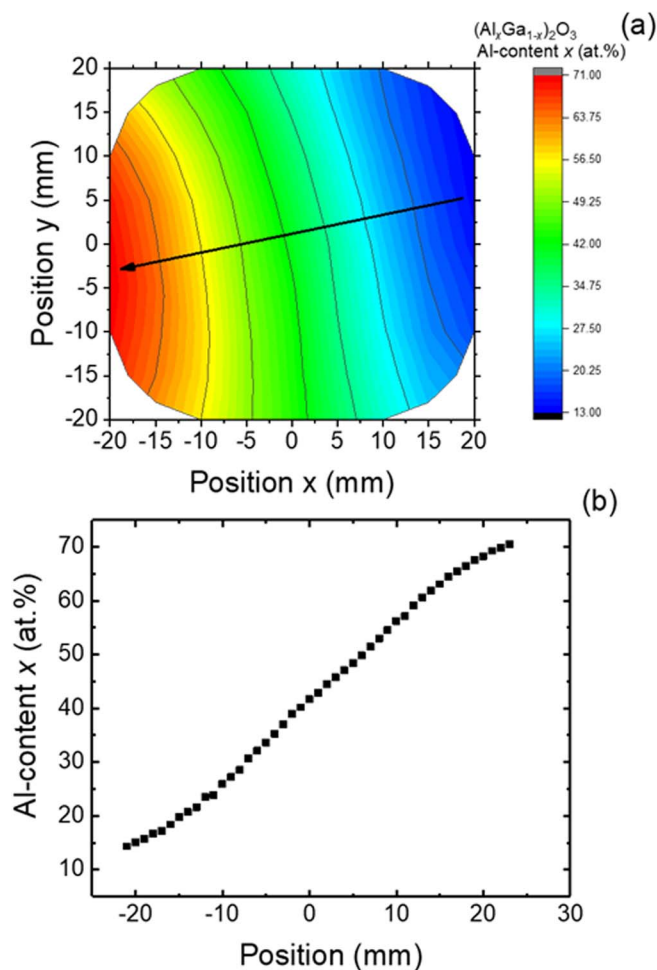

Figure 1. Typical (Al$_x$Ga$_{1-x}$)$_2$O$_3$/Ga$_2$O$_3$ HFET where gate insulator selection is crucial.

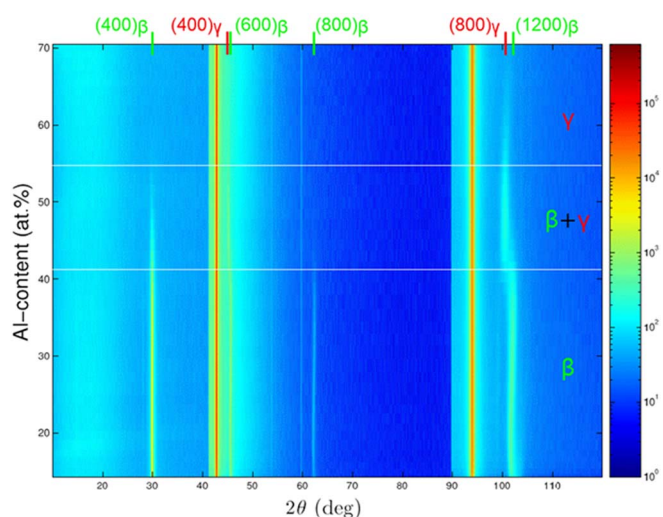*Electrochemical Society Student Member.
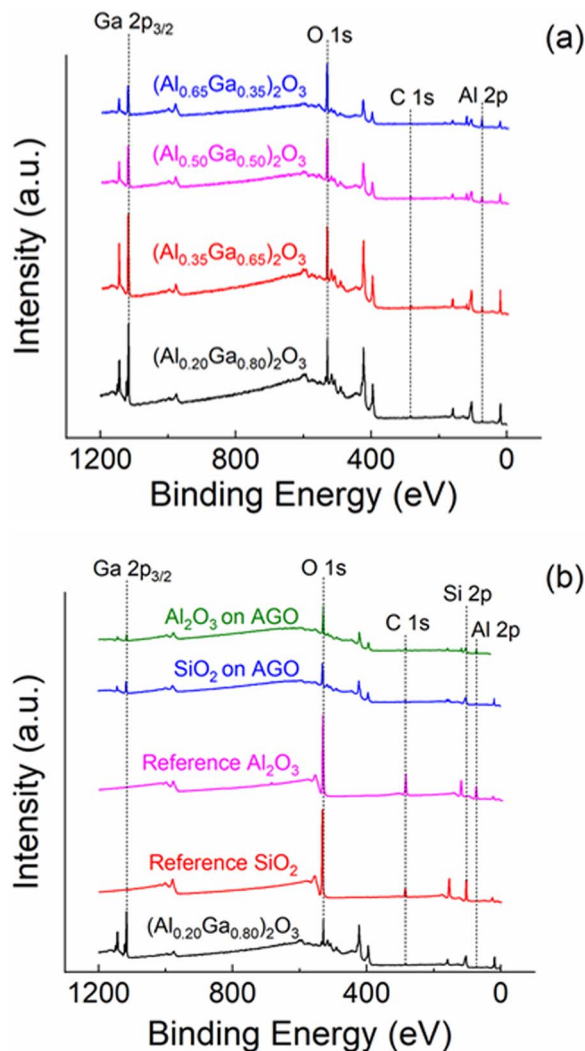**Electrochemical Society Fellow.
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Figure 2. (a) False-color representation of the Al concentration within a two inch in diameter $(Al_{1-x}Ga_x)_2O_3$ thin film grown with continuously varying composition on (100)MgO. (b) Line scan of Al content as a function of position along the wafer determined by EDX along the gradient direction depicted as black arrow in (a).

Figure 3. False-color representation of θ-2θ X-ray diffractograms acquired along the compositional gradient of a $(Al_xGa_{1-x})_2O_3$ CCS-PLD sample deposited on (100) MgO substrates deposited at about 650°C.

Figure 4. XPS survey scans of (a) $(Al_xGa_{1-x})_2O_3$ at the aluminum concentrations studied in this report and (b) thick ALD Al_2O_3, thick ALD SiO_2, and heterostructures of each oxide on AGO. The intensity is in arbitrary units (a.u.).

The lateral variation of the Al content of a $(Al_xGa_{1-x})_2O_3$ thin film grown by CCS-PLD using a target consisting of semicircular Al_2O_3 and Ga_2O_3 segments is shown in Figure 2a. Energy-dispersive X-ray spectroscopy (EDX) was used for the spatially resolved chemical analysis, where a Nova Nanolab 200 system by FEI company was employed. The sample was deposited at a growth temperature of 650°C and an oxygen pressure 0.08 mbar on a two inch in diameter (100) MgO substrate. The Al concentration varies between 0.15 and 0.70, as shown in Figure 2b and has a slight S-shaped dependence along the gradient direction, in agreement with calculations.[13] Along lines perpendicular to the gradient direction the Al concentration is in principle constant. Figure 3 shows a false color representation of X-ray diffractograms along the compositional gradient. As discussed above, there is pure monoclinic phase up to ∼40-45 at.%, then a mixed phase with γ-$(Al_xGa_{1-x})_2O_3$ is present. Above ∼50-55 at.%, the films show pure γ-phase. More details of the growth are given elsewhere.[6,13]
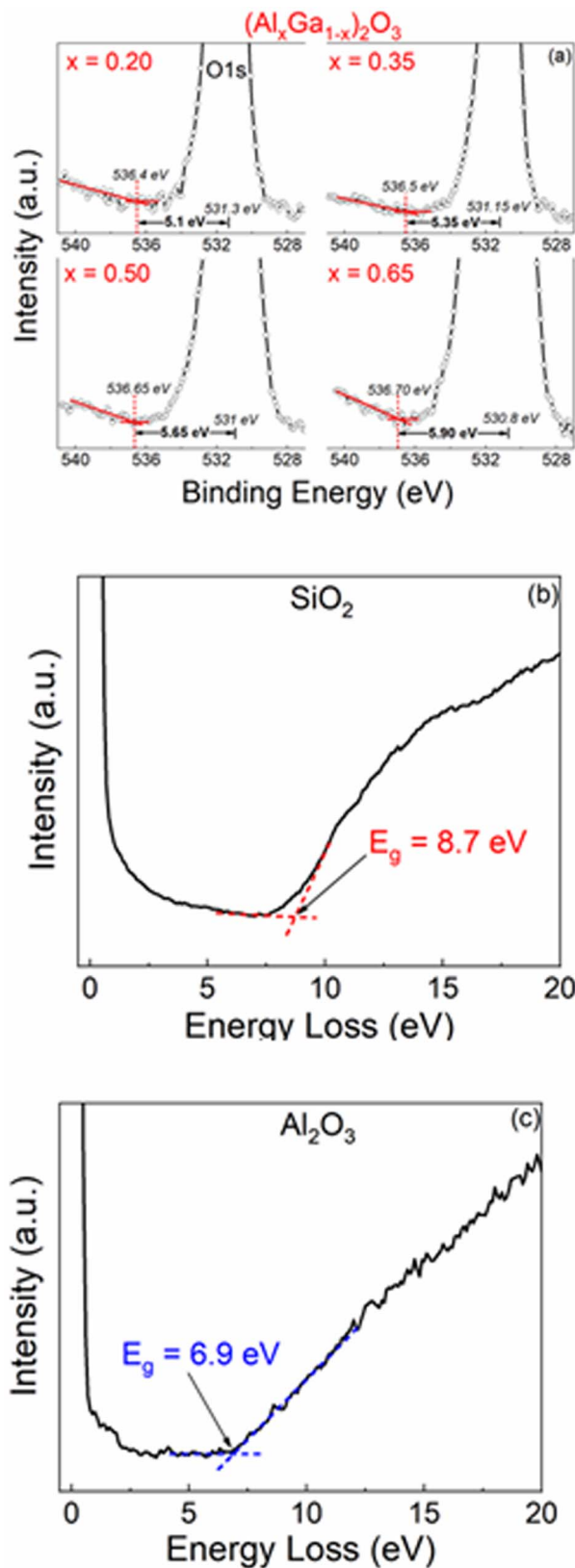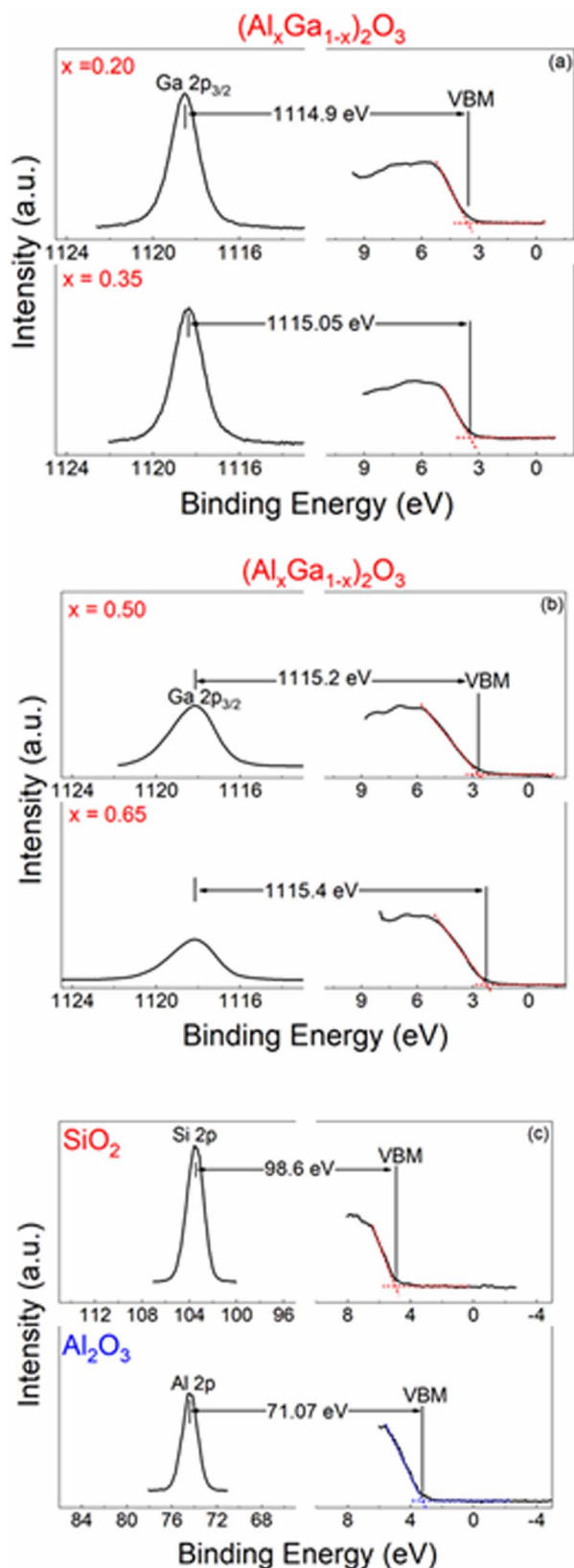
The ALD layers were deposited at 200°C in remote plasma mode in a Cambridge Nano Fiji 200 using a trimethylaluminum precursor or Tris (dimethylamino) silane and an inductively coupled plasma (ICP) at 300 W to generate atomic oxygen.[26–29] For substrate cleaning prior to deposition, a rinse sequence consisting of acetone and IPA was followed by drying in filtered N_2, and finally ozone exposure for 15 min. After this substrate cleaning, samples were directly loaded into the deposition systems within a cleanroom environment to avoid contamination of the deposited films. Both thick (200 nm) and thin

Figure 5. XPS spectra of core levels to valence band maximum (VBM) for (a) reference $(Al_xGa_{1-x})_2O_3$ with 20% and 35% Aluminum, (b) reference $(Al_xGa_{1-x})_2O_3$ with 50% and 65% Aluminum, and (c) ALD thick film Al_2O_3 and SiO_2. The intensity is in arbitrary units (a.u.).

Figure 6. Bandgap of (a) $(Al_xGa_{1-x})_2O_3$ determined using the onset of the plasmon loss feature in O 1s photoemission spectrum, (b) ALD SiO_2, and (c) ALD Al_2O_3 where both deposited films' bandgap was determined by Reflection Electron Energy Loss Spectra. The intensities are in arbitrary units (a.u.).

Table I. Summary of the measured reference and heterostructure peaks for SiO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ (eV).

Aluminum Concentration	Reference (Al$_x$Ga$_{1-x}$)$_2$O$_3$			Reference SiO$_2$			Thin SiO$_2$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$	
	Core Level Peak (Ga 2p$_{3/2}$)	VBM	Core - VBM	Core Level Peak (Si 2p)	VBM	Core - VBM	Δ Core Level (Ga 2p$_{3/2}$ - Si 2p)	Valence Band Offset
(Al$_{0.20}$Ga$_{0.80}$)$_2$O$_3$	1118.50	3.6	1114.90	103.40	4.80	98.60	1115.05	1.25
(Al$_{0.35}$Ga$_{0.65}$)$_2$O$_3$	1118.35	3.3	1115.05	-	-	-	1115.10	1.3
(Al$_{0.50}$Ga$_{0.50}$)$_2$O$_3$	1118.10	2.9	1115.20	-	-	-	1115.25	1.35
(Al$_{0.65}$Ga$_{0.35}$)$_2$O$_3$	1118.00	2.6	1115.40	-	-	-	1015.40	1.4

(1.5 nm) layers of the dielectrics were deposited for measuring both bandgaps and core levels on the β-(Al$_x$Ga$_{1-x}$)$_2$O$_3$.[30]

We used XPS survey scans to establish the chemical state of the SiO$_2$, Al$_2$O$_3$, and the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ samples. The XPS system was a Physical Instruments ULVAC PHI, with an Al X-ray source (energy 1486.6 eV, source power 300W), analysis size of 100 μm diameter, a take-off angle of 50° and acceptance angle of ± 7 degrees. The electron pass energy was 23.5 eV for high-resolution scans and 93.5 eV for survey scans. The total energy resolution of this XPS system is about 0.5 eV, and the accuracy of the observed binding energy is within 0.03 eV.[27–30]

To avoid sample charging, charge compensation employed an electron flood gun and a simultaneous ion beam. C 1s core levels of the surface adsorbate (284.8 eV) were used to calibrate the binding energy. Only the relative energy position is needed to determine the valence band offsets, so the absolute energy calibration for a sample has no effect on that number.[32,33] The samples were electrically insulated from the chuck to avoid uneven charge dispersion along the sample. All electron analyzers and equipment were grounded. Differential charging was not observed in any of the samples with the use of the electron gun. The SiO$_2$ and Al$_2$O$_3$ bandgaps were obtained from Reflection Electron Energy Loss Spectroscopy (REELS)[32,33] using a 1 kV electron beam and hemispherical electron analyzer. The bandgaps of the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ for each composition were obtained from XPS energy loss measurements of the O1S peak. This is conveniently done at the same time as the band alignment measurements.

Results and Discussion

The XPS survey scans from the different compositions of (Al$_x$Ga$_{1-x}$)$_2$O$_3$ are shown in Figure 4a. The samples show only the lattice constituents. Figure 4b shows the survey spectra for the thick (200 nm) dielectrics of ALD SiO$_2$ and Al$_2$O$_3$, thin (1.5 nm) SiO$_2$ and Al$_2$O$_3$ on β-(Al$_x$Ga$_{1-x}$)$_2$O$_3$, labelled here as AGO, and the (Al$_{0.2}$Ga$_{0.8}$)$_2$O$_3$ sample for reference.

Figure 5 shows high resolution XPS spectra for the vacuum-core delta regions of four different (Al$_x$Ga$_{1-x}$)$_2$O$_3$ compositions, namely x = 0.2 and 0.35 in (a), x = 50 and 0.65 in (b), along with the SiO$_2$ and Al$_2$O$_3$ in (c). The VBMs were 3.6 ± 0.2 eV for β-(Al$_{0.2}$Ga$_{0.8}$)$_2$O$_3$, 3.3 eV for (Al$_{0.35}$Ga$_{0.65}$)$_2$O$_3$, 2.9 eV for (Al$_{0.5}$Ga$_{0.5}$)$_2$O and 2.6 eV for (Al$_{0.65}$Ga$_{0.35}$)$_2$O$_3$. The valence band offsets are then obtained by measuring the shift of the core levels for the heterostructure samples with the thin dielectric on top of the different compositions of (Al$_x$Ga$_{1-x}$)$_2$O$_3$.[34] We also measured the bandgaps of the

(Al$_x$Ga$_{1-x}$)$_2$O$_3$ at the compositions of interest, as shown in Figure 6a, from the separation between the core level peak energy and the onset of inelastic (plasmon) losses in each O 1s photoemission spectra.[35] The respective bandgaps were 5.1 eV for (Al$_{0.2}$Ga$_{0.8}$)$_2$O$_3$, 5.35eV for (Al$_{0.35}$Ga$_{0.65}$)$_2$O$_3$, 5.65 eV for (Al$_{0.5}$Ga$_{0.5}$)$_2$O$_3$ and 5.90 eV for (Al$_{0.65}$Ga$_{0.35}$)$_2$O$_3$. These are in excellent agreement with the relationship reported previously for the compositional dependence of bandgap (E$_g$) of (Al$_x$Ga$_{1-x}$)$_2$O$_3$, namely[1,7,36]

$$E_g = (4.75 + 1.87x) \, \text{eV}$$

Using this relationship, we would expect values of 5.1eV for x = 0.2, 5.4 eV for x = 0.35, 5.68 eV for x = 0.50 and 5.97 eV for x = 0.65., ie. the differences from our experimental values are <0.07 eV across the composition range studied here. Values of the indirect bandgap using the formula E$_g$ = 4.637 +1.87x, determined by Schmidt-Grund et al.[7] on a similar CCS-PLD sample by using spectroscopic ellipsometry, are 5.01 eV, 5.29 eV, 5.57 eV and 5.85 eV for x = 0.2, 0.35, 0.5 and 0.65, respectively, which is in good agreement to the values determined by XPS. Other experimental values for similar compositions reported from studies by Feng et al.[25,26] include 5.1 eV (x = 0.35), 5.3eV (x = 0.33), 5.43 eV (x = 0.30), 5.2eV (x = 0.40), 5.64 eV (x = 0.49), and 5.4 eV (x = 0.53). Obviously, within similar groups of samples in those cases, there was variation of ~0.2 eV. Wakabayashi et al.[20] reported that strain in layers of (Al$_x$Ga$_{1-x}$)$_2$O$_3$ might lead to bowing of the bandgap with composition. Figures 6b and 6c shows the REELS spectra to determine the bandgap of the SiO$_2$ and Al$_2$O$_3$, respectively, with values of 8.7 eV and 6.9 eV, respectively. These are consistent with previous reported values.[26–30]

Tables I and II show the valence band maximum (VBM) for the dielectrics and the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ obtained using linear fitting of the leading edge of the valence band. Figure 7 shows the core energy differences from XPS spectra for (Al$_x$Ga$_{1-x}$)$_2$O$_3$ to SiO$_2$ for compositions of 0.20 and 0.35 (a) and 0.50 and 0.65 (b), respectively, as well as (Al$_x$Ga$_{1-x}$)$_2$O$_3$ to Al$_2$O$_3$ for compositions of 0.20 and 0.35 (c) and 0.50 and 0.65 (d), respectively. The core energy levels and the differences between Ga 2p3/2 and Si 2p or Al 2p core energy levels, respectively, are shown in the figure. We used the usual method to measure the valence band offsets by observing the shift of the core levels from the (Al$_x$Ga$_{1-x}$)$_2$O$_3$ when SiO$_2$ or Al$_2$O$_3$ was deposited.[35] This method measures the energy difference between a core level and the VBM for both the single layer dielectric and (Al$_x$Ga$_{1-x}$)$_2$O$_3$. The separation between the reference core levels can be translated into the valence band offset (VBO) using the previously measured single layer sample core-level to valence band maximum (VBM) energies.[35] The VBM

Table II. Summary of the measured reference and heterostructure peaks for Al$_2$O$_3$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$ (eV).

| Aluminum Concentration	Reference (Al$_x$Ga$_{1-x}$)$_2$O$_3$			Reference Al$_2$O$_3$			Thin Al$_2$O$_3$ on (Al$_x$Ga$_{1-x}$)$_2$O$_3$	
	Core Level Peak (Ga 2p$_{3/2}$)	VBM	Core - VBM	Core Level Peak (Al 2p)	VBM	Core - VBM	Δ Core Level (Ga 2p$_{3/2}$ - Al 2p)	Valence Band Offset
(Al$_{0.20}$Ga$_{0.80}$)$_2$O$_3$	1118.50	3.6	1114.90	74.32	3.25	71.07	1043.60	0.23
(Al$_{0.35}$Ga$_{0.65}$)$_2$O$_3$	1118.35	3.3	1115.05	-	-	-	1043.70	0.28
(Al$_{0.50}$Ga$_{0.50}$)$_2$O$_3$	1118.10	2.9	1115.20	-	-	-	1043.80	0.33
(Al$_{0.65}$Ga$_{0.35}$)$_2$O$_3$	1118.00	2.6	1115.40	-	-	-	1044.00	0.33
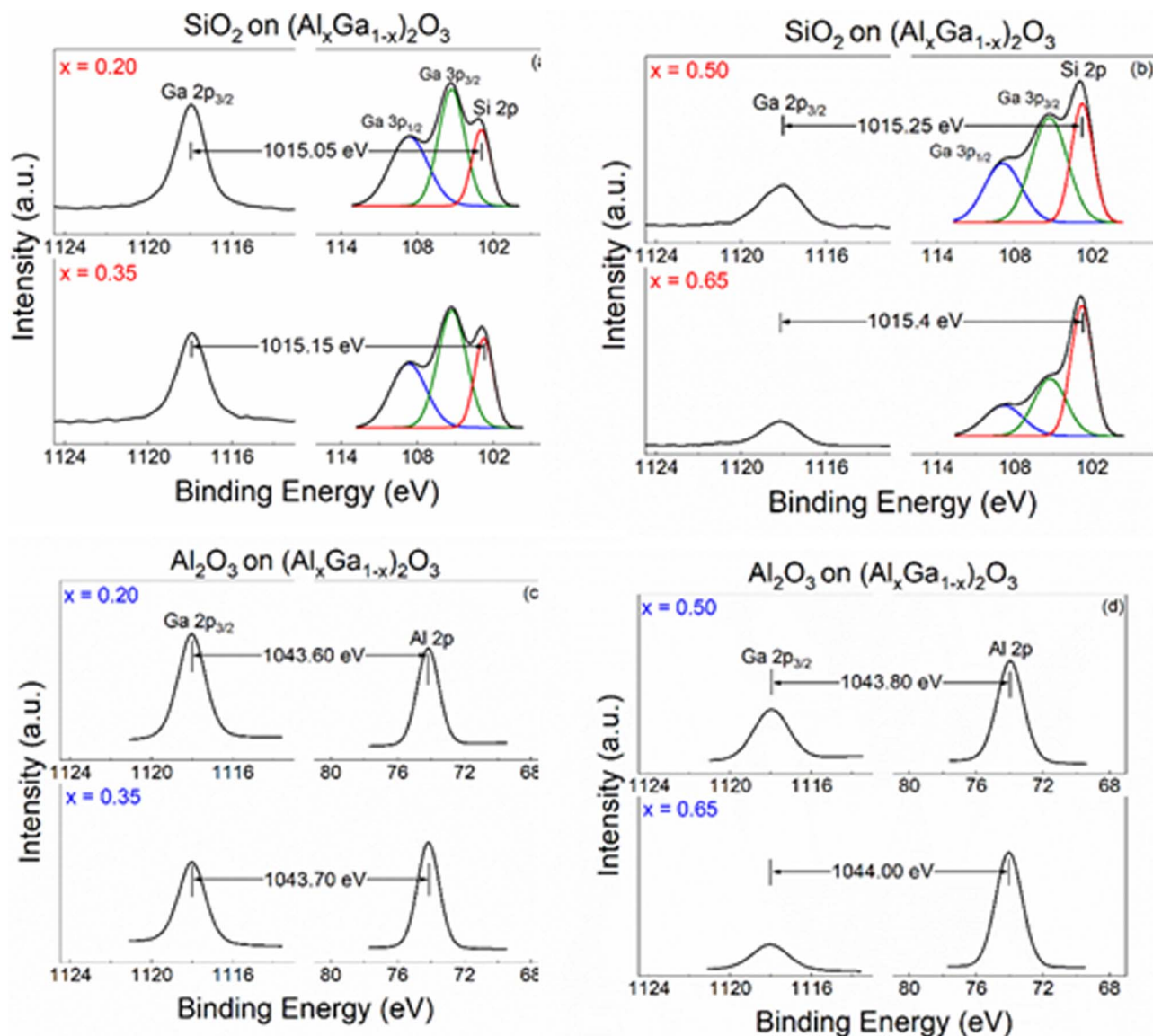
Figure 7. High resolution XPS spectra for the (a-b.) $(Al_xGa_{1-x})_2O_3$ to SiO_2 core delta regions and the (c-d.) $(Al_xGa_{1-x})_2O_3$ to Al_2O_3 core delta regions. The intensity is in arbitrary units (a.u.).

values were determined by linear extrapolation of the leading edge to the baseline of the valence band spectra. The error bars in the different binding energies were combined in a root sum square relationship to determine the overall error bars in the valence band offsets.[30]

The valence band offsets for SiO_2 were 1.25 ± 0.20 eV for β-$(Al_{0.2}Ga_{0.8})_2O_3$, 1.3 ± 0.20 eV for $(Al_{0.35}Ga_{0.65})_2O_3$, 1.35 ± 0.20 eV for $(Al_{0.5}Ga_{0.5})_2O$ and 1.4 ± 0.20 eV for $(Al_{0.65}Ga_{0.35})_2O_3$. Based on the measured bandgap of this dielectric, the conduction band offsets are then 2.35 eV (x = 0.2), 2.20 eV (x = 0.35), 1.7 eV (x = 0.4) and 1.4 eV (x = 0.65). SiO_2 therefore provides excellent confinement of electrons in $(Al_xGa_{01-x})_2O_3$ samples over the practical range of Al contents achievable. The $SiO_2/β$-$(Al_xGa_{1-x})_2O_3$ band alignment remained type I across the entire composition range examined here, as shown in the schematic of Figure 8. Note that our valence band offsets are 0.10-0.45 eV different (both larger or smaller, depending on composition) than reported by Feng et al.[24,25] for similar deposition conditions for the SiO_2 on $(Al_xGa_{01-x})_2O_3$ of comparable compositions to those used here. There are no obvious signs of metal contamination in the reported XPS survey spectra, so this gives an idea of the inherent accuracy of comparing valence band offsets values in the literature for the same dielectric/semiconductor systems.
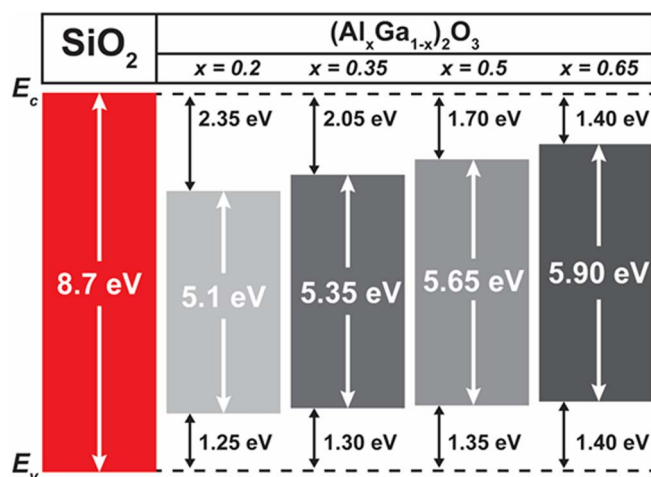
Figure 8. Band diagrams for the $SiO_2/(Al_xGa_{1-x})_2O_3$ heterostructure in which the SiO_2 was deposited by ALD.
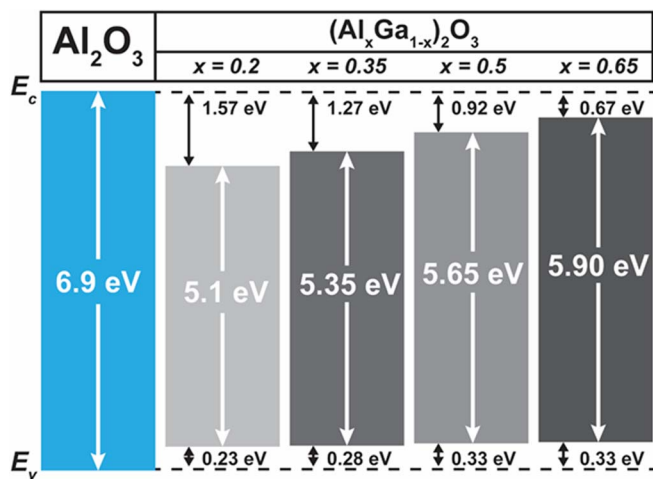
Figure 9. Band diagrams for the $Al_2O_3/(Al_xGa_{1-x})_2O_3$ heterostructure in which the Al_2O_3 was deposited by ALD.

The valence band offsets for Al_2O_3 were 0.23 ± 0.05 eV for β-$(Al_{0.2}Ga_{0.8})_2O_3$, 0.28 ± 0.05 eV for $(Al_{0.35}Ga_{0.65})_2O_3$, 0.33 ± 0.06 eV for $(Al_{0.5}Ga_{0.5})_2O$ and 0.33 ± 0.06 eV for $(Al_{0.65}Ga_{0.35})_2O_3$. Based on the measured bandgap of Al_2O_3, the conduction band offsets are then 1.57 eV (x = 0.2), 1.27 eV (x = 0.35), 0.92 eV (x = 0.4) and 0.67 eV (x = 0.65). The electron confinement would be marginal at high Al contents in $Al_2O_3/(Al_xGa_{1-x})_2O_3$. The band alignments are also type I, as shown in the schematic of Figure 9.

Conclusions

XPS was used to measure the valence band offsets of $SiO_2/(Al_xGa_{01-x})_2O_3$ and $Al_2O_3/(Al_xGa_{01-x})_2O_3$ heterojunctions over the widest range of Al contents reported to date (x = 0.2-0.65), in which the dielectrics were deposited by ALD. The band alignments are type I in all cases, with valence band offsets >1.25 eV for SiO_2 across the whole composition range of $(Al_xGa_{1-x})_2O_3$ examined. By contrast, the valence band offsets for Al_2O_3 are in the range 0.23-0.33 eV for the same range of Al contents in $(Al_xGa_{1-x})_2O_3$. The CCS-PLD technique provides an effective pathway to producing a wide range of compositions for study of band alignments.

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