

# Al Composition Dependence of Band Offsets for SiO<sub>2</sub> on $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub>

To cite this article: Xinyi Xia et al 2021 ECS J. Solid State Sci. Technol. 10 113007

View the article online for updates and enhancements.



### **ECS** Membership = Connection

#### ECS membership connects you to the electrochemical community:

Visit electrochem.org/join

 Facilitate your research and discovery through ECS meetings which convene scientists from around the world;

- Access professional support through your lifetime career:
- Open up mentorship opportunities across the stages of your career;
- Build relationships that nurture partnership, teamwork—and success!

ECS

This content was downloaded from IP address 128.227.144.107 on 08/08/2022 at 16:35

Join ECS!





## Al Composition Dependence of Band Offsets for SiO<sub>2</sub> on $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub>

Xinyi Xia,<sup>1</sup> Chaker Fares,<sup>1</sup> Fan Ren,<sup>1</sup> Anna Hassa,<sup>2</sup> Holger von Wenckstern,<sup>2</sup> Marius Grundmann,<sup>2</sup> and S. J. Pearton<sup>3,z</sup>

<sup>1</sup>Department of Chemical Engineering, University of Florida, Gainesville FL 32611 United States of America <sup>2</sup>Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, 04103 Leipzig, Germany <sup>3</sup>Department of Materials Science and Engineering, University of Florida, Gainesville FL 32611 United States of America

Valence band offsets for SiO<sub>2</sub> deposited by Atomic Layer Deposition on  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> alloys with x = 0.26-0.74 were measured by X-ray Photoelectron Spectroscopy. The samples were grown with a continuous composition spread to enable investigations of the band alignment as a function of the alloy composition. From measurement of the core levels in the alloys, the bandgaps were determined to range from 5.8 eV (x = 0.26) to 7 eV (x = 0.74). These are consistent with previous measurements by transmission spectroscopy. The valence band offsets of SiO<sub>2</sub> with these alloys of different composition were, respectively, were -1.2 eV for x = 0.26, -0.2 eV for x = 0.42, 0.2 eV for x = 0.58 and 0.4 eV for x = 0.74. All of these band offsets are too low for most device applications. Given the bandgap of the SiO<sub>2</sub> was 8.7 eV, this led to conduction band offsets of 4.1 eV (x = 0.26) to 1.3 eV (x = 0.74). The band alignments were of the desired nested configuration for x > 0.5, but at lower Al contents the conduction band offsets were negative, with a staggered band alignment. This shows the challenge of finding appropriate dielectrics for this ultra-wide bandgap semiconductor system.

© 2021 The Electrochemical Society ("ECS"). Published on behalf of ECS by IOP Publishing Limited. [DOI: 10.1149/2162-8777/ ac39a8]

Manuscript submitted September 18, 2021; revised manuscript received November 8, 2021. Published November 25, 2021.

 $(Al_xGa_{1-x})_2O_3$  is an ultra-wide bandgap semiconductor that is attracting attention for use in high power transistors and deep ultraviolet (DUV) photodetectors.<sup>1-4</sup> To this point, most of the work has focused on the  $\beta$ -polytype with a bandgap tunability ranging from 4.8 eV (Ga<sub>2</sub>O<sub>3</sub>) to 8.7 eV (Al<sub>2</sub>O<sub>3</sub>).<sup>1,2</sup> The advantages of  $\beta$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> grown on  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> include the high channel mobility at the interface and ability to use the heterostructure in field effect transistor structures with improved mobility, as well as Schottky diodes and photodetectors.<sup>1,2</sup> A drawback of this materials system during growth with techniques such as Metal Organic Chemical Vapor Deposition is the difficulty in maintain phase stability at high Al contents.<sup>1</sup>

The  $\alpha$ -polytype is a potential solution to extending the stability to higher Al contents and higher temperatures.<sup>2-12</sup> This metastable, corundum crystal structure  $\alpha$ -polytype, transforms to the  $\beta$ -polymorph at temperatures above 750 °C-900 °C under equilibrium conditions.<sup>5</sup> Of all reported polymorphs, this phase has the highest optical bandgap with energies ranging between 5.0 eV and 5.3 eV for the binary. A number of different growth methods have recently been shown to produce high quality rhombohedral  $\alpha$ -Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> over a broad composition range<sup>6-48</sup> and in particular, the use of  $Al_2O_3$  cap layers during annealing is able to preserve the  $\alpha$ -phase.<sup>16</sup> The  $\alpha$ -polytype has an even larger bandgap than the  $\beta$ -polytype and can be grown on low-cost c-plane sapphire substrates which have the same lattice structure, albeit with relatively high dislocation densities.<sup>2,19,22,43</sup> Figure 1 shows the crystal structure of rhombohedral  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>. This rhombohedral phase can also occur for Al<sub>2</sub>O<sub>3</sub> and  $In_2O_3$ , which allows a potential bandgap engineering over the entire composition range,<sup>34,45,46</sup> rendering this polymorph interesting for deep UV photodetectors or quantum-well infrared photodetectors,<sup>1,2</sup> in addition to the power device applications.<sup>35–37</sup>

It is clearly of interest to establish the band alignments for commonly employed dielectrics on this materials system. SiO<sub>2</sub> is one of the most promising choices, due to its large bandgap and maturity as a dielectric in semiconductor devices. We have previously measured the band alignment and its thermal stability for SiO<sub>2</sub> and Al<sub>2</sub>O on  $\beta$ - (Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> for 0.2 < x < 0.65.<sup>49,50</sup> In this paper we report on the measurement of valence band offsets in the SiO<sub>2</sub>/ $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> system using alloys grown by Pulsed Laser Deposition (PLD)<sup>51,52</sup> over a wide composition range. This was done using circular half-segmented (Ga<sub>2</sub>O<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub>) ceramic targets to synthesize large diameter large thin films with a lateral varying cation composition.<sup>1,7,8,45,46</sup> Since the bandgap of the  $\alpha$ -polytype alloys is even larger than those of the  $\beta$ -polytype alloys, this system represents a test in terms of finding dielectrics that exhibit acceptable band offsets (>1 eV is a common rule of thumb) in both the conduction and valence bands.

Since Schottky barrier diodes are expected to be one of the first commercial applications of Ga<sub>2</sub>O<sub>3</sub>, having an understanding of band alignments for dielectrics on this materials system is key in designing edge termination schemes to avoid field crowding at the device contact periphery. In addition, the reliability of the dielectric layer in these devices and also Metal-Insulator-Semiconductor structures under very high electric field is not established. As discussed above, both dielectric/semiconductor and the gate/dielectric interface should have large band offsets. If the interface has a small conduction band offset, application of high electric fields (typically >10<sup>7</sup> V. cm<sup>-1</sup>) will cause carrier injection via Fowler-Nordheim tunneling.<sup>53,54</sup> This can lead to breakdown of the dielectric. A small conduction band offset will also produce carrier injection into the dielectric and Poole-Frenkel conduction within the dielectric.<sup>53,54</sup> Similarly, small valence band offsets can produce higher gate leakage during exposure to deep-UV illumination.

#### Experimental

The  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> alloys were grown by combinatorial PLD using a KrF excimer laser (Coherent LPX Pro 305, 248 nm, 2.6 J.cm<sup>-2</sup>) directed through a quartz window onto a segmented ceramic target (Ga<sub>2</sub>O<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub>) under an O<sub>2</sub> partial pressure.<sup>1,51,52</sup> The substrate and target were mounted rotatable opposite each other at a distance of 10 cm and with a small lateral offset to each other.<sup>2</sup> PLD has been utilized previously to obtain AGO alloys over a broad composition range for both  $\alpha$  and  $\beta$ -polytypes<sup>8,20,40</sup> The resulting cation ratio was determined by energy dispersive X-ray spectroscopy (EDX) measurements performed using a FEI Nova Nanolab 200 equipped with an Ametek EDAX detector and the crystal structure by X-ray diffraction (XRD). measured by a PANalytical X'pert PRO MRD diffractometer equipped with a PIXcel3D detector operating in 1D scanning line mode with 255 channels (for  $2\theta$ - $\omega$  scans), receiving slit mode (for  $\Phi$ -scans) and fast 2D frame- based mode (for reciprocal space maps). The cation gradient ranged between x =0.13 and x = 0.84 as measured by EDX and marked as black dots in Fig. 2a. The composition between these data points was interpolated and the resulting cation ratio represented as false color map. Along the cation gradient direction, marked with a black arrow, additional



Figure 1. Crystal structure of rhombohedral  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>. The oxygen atoms are displayed. as red and the Ga atoms as green spheres. The graphic was created with the software Crystal Maker X, http://www.crystalmaker.com/about/index.html.

EDX measurements were performed in 1 mm steps to obtain the spatial Al dependence with high lateral resolution as shown in Fig. 2b.<sup>20</sup> The Al incorporation covers a range of  $0.13 \le x \le 0.84$ . The rhombohedral crystal structure of the thin films was confirmed by the  $2\theta$ - $\omega$  XRD scans. The resulting false color map is presented in Fig. 3 and reveals the crystallization in the rhombohedral phase in the entire investigated Al range. We examined four compositions, x = 0.26, 0.42, 0.58 and 0.74. More details and discussion on the characterization of these samples can be found in Hassa et al.<sup>20</sup>

The SiO<sub>2</sub> layers were deposited by Atomic Layer Deposition (ALD) at 200 °C using the thermal mode in a Cambridge Nano Fiji 200. The precursors were Tris (dimethylamino) silane and an inductively coupled plasma (ICP) of O<sub>2</sub> at 300 W.<sup>49,50</sup> After solvent cleaning, the substrates were exposed to Ozone for 15 min to form a protective oxide that is easily thermally desorbed during the ALD step. Both thick (200 nm) and thin (1.5 nm) layers of SiO<sub>2</sub> were deposited for measuring bandgaps and core levels on the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub>.

XPS survey scans were performed to measure the chemical state of the SiO<sub>2</sub> and (Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> layers in a Physical Instruments ULVAC PHI, with an Al X-ray source (energy 1486.6 eV, source power 300 W).<sup>49,50,53,55</sup> The analysis size was 100  $\mu$ m diameter, with 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 0.5 eV, and the accuracy of the observed binding energy is ~0.03 eV. Charge compensation was performed using an electron flood gun and simultaneous ion beam. The C 1 s core levels of the surface adsorbate (284.8 eV) were used to calibrate the binding energy.<sup>55</sup> Only the relative energy position is needed to determine the valence band offsets, so the absolute energy calibration has no effect on that offset. 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.<sup>49,50</sup>

The SiO<sub>2</sub> bandgap was obtained from Reflection Electron Energy Loss Spectroscopy (REELS) using a 1 kV electron beam and hemispherical electron analyzer. The bandgaps of the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> for each composition were obtained from XPS energy loss measurements of the O1S peak. This is done simultaneously with the band alignment measurements.<sup>50,55</sup>

#### **Results and Discussion**

Figure 4 shows the high resolution XPS spectra for the  $\alpha$ - (Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> to SiO<sub>2</sub> vacuum-core delta regions of the four



Figure 2. (a) Al content x at 49 points across the thin film surface, marked on the map as black dots and determined by EDX. The data between the measurement points was interpolated, the black arrow represents the direction of the gradient. (b) Al ratio x acquired along the indicated gradient in (a) by EDX and XPS, respectively.

compositions, while the data for the SiO<sub>2</sub> is shown in Fig. 5. The valence band offsets were extracted from the shift of the core levels for the heterostructure samples with the thin dielectric on top of the four different compositions of the alloy. A compilation of the valence band maxima (VBM) is collected in Table I, with values of  $(2.7 \pm 0.15)$  eV for x = 0.26,  $(3.3 \pm 0.15)$  eV for x = 0.42,  $(4.4 \pm 0.15)$ eV for x = 0.58 and  $(4.5 \pm 0.15)$  eV for x = 0.74. These were obtained by fitting of the leading edge of the valence band. 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  $(\Delta E_V)$ .<sup>49</sup> These band offsets are then obtained by measuring the shift of the core levels in the  $\alpha$ -  $(Al_xGa_{1-x})_2O_3$  when SiO<sub>2</sub> was deposited. These are also tabulated in Table I, with values

of -1.2 eV for x = 0.26, -0.2 eV for x = 0.42, 0.2 eV for x = 0.58and 0.4 eV for x = 0.74. Note the bandgap values derived from the XPS data are in excellent agreement with previous values obtained using UV/VIS dual beam spectrometry.<sup>8,9,20</sup>

Once the valence band offsets are established, to determine the conduction band offset, it is necessary to measure the band gap of each composition. We measured the bandgaps of the four  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> compositions, as shown in Fig. 6a, from the separation between the core level peak energy and the onset of inelastic (plasmon) losses in each O 1s photoemission spectra.<sup>36</sup> To find the band-gap energy, a linear fit is made to the measured loss spectra curve near the approximate location of onset of inelastic losses. Next, by subtracting the background, the "zero" level is



Figure 4. XPS spectra of core levels to valence band maximum (VBM) for reference  $(Al_xGa_{1-x})_2O_3$  with x = 0.26, 0.42, 0.58 or 0.74 (top to bottom) Aluminum, and (c) ALD thick film  $Al_2O_3$  and  $SiO_2$ . The intensity is in arbitrary units (a.u.).

Aluminum Concentration	Reference $(Al_xGa_{1-x})_2O_3$			Reference SiO <sub>2</sub>			Thin SiO <sub>2</sub> on $(Al_xGa_{1-x})_2O_3$	
	Core Level Peak (Ga 2p <sub>3/2</sub> )	VBM	Core - VBM	Core Level Peak (Si 2p)	VBM	Core -VBM	$\Delta$ Core Level (Ga 2p <sub>3/2</sub> - Si 2p)	Valence Band Offset
(Al <sub>0.26</sub> Ga <sub>0.74</sub> ) <sub>2</sub> O <sub>3</sub>	1117.7	$2.7 \pm 0.15$	1115	103.40	4.80	98.60	1017.6	-1.2
(Al <sub>0.42</sub> Ga <sub>0.58</sub> ) <sub>2</sub> O <sub>3</sub>	1118.2	$3.3 \pm 0.15$	1114.9		_	_	1016.5	-0.2
(Al <sub>0.58</sub> Ga <sub>0.42</sub> ) <sub>2</sub> O <sub>3</sub>	1119.2	$4.4 \pm 0.15$	1114.7		_	_	1015.9	0.2
$(Al_{0.74}Ga_{0.26})_2O_3$	1118.9	$4.5 \pm 0.15$	1114.5	_	_		1015.5	0.4

Table I. Summary of the measured reference and heterostructure peaks for SiO<sub>2</sub> on (Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> (eV).



Figure 5. XPS spectra of core levels to valence band maximum (VBM) for ALD thick film SiO2. The intensity is in arbitrary units (a.u.).



Figure 6. Bandgap of  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> determined using the onset of the plasmon loss feature in O 1 s photoemission spectrum. The intensities are in arbitrary units (a.u.).

determined. The energy corresponding to the onset of inelastic losses is found by extrapolating the linear-fit line and calculating its intersection with the "zero" level.<sup>33,40</sup> The bandgap energy is equal to the difference between the core-level peak energy and the onset of inelastic losses.

The bandgap energies were determined as 5.8 eV for x = 0.26, 6.1 eV for x = 0.42, 6.4 eV for x = 0.58 and 7 eV for x = 0.74, respectively. Figure 7 shows the relationship between bandgap of the alloys and Al composition for our samples. As previously reported, the bandgap as a function of composition, x, is given by<sup>28,56</sup>



**Figure 7.** Bandgap of  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> for 4 Al compositions (experimental points) fitted to a bowing parameter of 3 eV, along with a theoretical curve with bowing parameter 1.37 eV.

$$E_G(x) = (1 - x)E_G^{GaO} + xE_G^{AlO} - bx (1 - x)$$

where  $E_G^{GaO}$  and  $E_G^{AIO}$  are the bandgaps of the  $\alpha$ -polytype binary endpoints and b is the bowing parameter. This has been previously reported in the range 0.84–2.13.<sup>9,34</sup> We get a value closer to 3 for our samples in the composition range measured. It was noted by Varley<sup>56</sup> that the *b* values are strongly influenced by the selective occupation of different cation sublattice by Al and that a single bowing parameter is not accurate over the entire composition range

The SiO<sub>2</sub> bandgap was obtained from Reflection Electron Energy Loss Spectroscopy (REELS) and showed a value of 8.7 eV, similar to previous reports.<sup>49,50</sup>

The conduction band offsets  $\Delta E_{C}$  were obtained from the relation

$$\Delta E_C = E_G^{SiO2} - E_G^{GaO} - \Delta E_V$$

The corresponding conduction band offsets were 4.1 eV (x = 0.26), 2.8 eV (x = 0.46), 2.1 eV (x = 0.58) and 1.30 eV (x = 0.74).

Figure 8 shows the band diagrams for the SiO<sub>2</sub>/ $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> heterostructure. The SiO<sub>2</sub> has adequate conduction band offsets, but the valence band offsets are small and actually negative at x < 0.5. These would be inadequate confinement of holes in SiO<sub>2</sub>/ $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> samples over the entire composition range. The conduction band offsets are high enough to provide effective electron confinement. The SiO<sub>2</sub>/ $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> band alignment is type I for x > 0.5 and staggered for x < 0.5. It will be interesting to measure the band alignments for Al<sub>2</sub>O<sub>3</sub> deposited on the  $\alpha$ - (Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> alloys, since that is one of the few dielectrics with a high enough bandgap to provide effective carrier confinement for electrons and holes in these alloys.



Figure 8. Band diagrams for the  $SiO_2/\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> heterostructure in which the  $SiO_2$  was deposited by ALD.

#### **Summary and Conclusions**

The valence band offsets of  $SiO_2/\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> heterojunctions were measured over a range of Al contents (x = 0.26 - 0.74). The band alignments are staggered type II for x < 0.5 and straddling type I for x > 0.5, with conduction band offsets >1.3 eV across the composition range examined. The thermal stability of this system is of interest for future work, as the  $\alpha$ -polytype I not the most energetically favorable state, although kinetically limited process such as rapid thermal annealing will be able to be used in device processing steps without major issues. The conduction band offsets are sufficient for majority carrier devices such as MIS or Schottky diodes but the valence band offsets are too small or the wrong sign for effective confinement of holes and thus will not be useful for photodetectors fabricated on these Al-based alloys. The use of integration of higher-K gate dielectrics with large conduction band offset could be used to increase gate capacitance and thereby increase the operating frequency of heterostructure transistors based on AGO alloys. Additionally, high permittivity gate insulators are necessary to increase the vertical breakdown strength normal to the channel in lateral transistor structures. Thus, the search for high-K dielectrics with large and offsets to  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> remains as a challenge because while SiO<sub>2</sub> has sufficient conduction band offsets, it has a low dielectric constant.

#### Acknowledgments

The work was performed as part of Interaction of Ionizing Radiation with Matter University Research Alliance (IIRM-URA), sponsored by the Department of the Defense, Defense Threat Reduction Agency under award HDTRA1-20-2-0002. The content of the information does not necessarily reflect the position or the policy of the federal government, and no official endorsement should be inferred. The work at UF was also supported by NSF DMR 1856662 (James Edgar). The authors at Leipzig thank Jörg Lenzner for EDX measurements and Monika Hahn for PLD target preparation. A.H. acknowledges support by the European Social Fund within the Young Investigator Group "Oxide Heterostructures" (SAB 100310460) and the Leipzig School for Natural Sciences BuildMoNa.

#### ORCID

S. J. Pearton (D) https://orcid.org/0000-0001-6498-1256

#### References

- 1. E. Ahmadi and Y. Oshima, J. Appl. Phys., 126, 160901 (2019).
- 2. H. von WencksternGroup-III Sesquioxides: Growth, Physical Properties and Devices, Advanced Electronic Materials, 3, 1600350 (2017).
- 3. T. Kawaharamura, G. T. Dang, and M. Furuta, Jpn. J. Appl. Phys., 51, 040207 (2012).
- 4. Y. Oshima, E. G. Víllora, and K. Shimamura, Appl. Phys. Express, 8, 055501 (2015).
- 5. D. Machon, P. F. McMillan, B. Xu, and J. Dong, Phys. Rev. B, 73, 094125 (2006).
- 6. D. Shinohara and S. Fujita, Jpn. J. Appl. Phys., 47, 7311 (2008). 7. A. Hassa, C. Sturm, M. Kneiß, D. Splith, H. von Wenckstern, T. Schultz, N. Koch,
- M. Lorenz, and M. Grundmann, APL Mater., 8, 021103 (2020). 8. A. Hassa, C. Wouters, M. Kneiß, D. Splith, C. Sturm, H. von Wenckstern,
- M. Albrecht, M. Lorenz, and M. Grundmann, J. Phys D: Applied Physics, 53, 485105 (2020). 9. R. Jinno et al., Sci. Adv., 7, eabd5891 (2021).
- 10. A. F. M. Anhar Uddin Bhuiyan, Z. Feng, L. Meng, and H. Zhao, J. Mat Res (2021), in press.
- 11. M. Lorenz, S. Hohenberger, E. Rose, and M. Grundmann, Appl. Phys. Lett., 113, 231902 (2018).
- 12. M. Hilfiker, U. Kilic, M. Stokey, R. Jinno, Y. Cho, H. G. Xing, D. Jena, R. Korlacki, and M. Schubert, Appl. Phys. Lett., 119, 092103 (2021).
- 13. R. Jinno, T. Uchida, K. Kaneko, and S. Fujita, Appl. Phys. Express, 9, 071101 (2016).

- 14. S. Fujita and K. Kaneko, J. Cryst. Growth, 401, 588 (2014).
- 15. K. Kaneko, K. Suzuki, Y. Ito, and S. Fujita, J. Cryst. Growth, 436, 150 (2016).
- 16. J. P. McCandless et al., Appl. Phys. Lett., 119, 062102 (2021).
- S. T. Piccandoss et al., *Appl. Phys.*, **51**, 070203 (2012).
   K. Akaiwa and S. Fujita, *Jpn. J. Appl. Phys.*, **51**, 070203 (2012).
   M. Oda, R. Tokuda, H. Kambara, T. Tanikawa, T. Sasaki, and T. Hitora, *Appl.* Phys. Express, 9, 021101 (2016).
- 19. K. Akaiwa, K. Kaneko, K. Ichino, and S. Fujita, Jpn. J. Appl. Phys., 55, 1202BA (2016).
- 20. A. Hassa, P. Storm, M. Kneiß, D. Splith, H. von Wenckstern, M. Lorenz, and M. Grundmann, Phys. Status Solidi B, 258, 2000394 (2020).
- 21. J. E. N. Swallow, R. G. Palgrave, P. A. E. Murgatroyd, A. Regoutz, M. Lorenz, A. Hassa, M. Grundmann, H. von Wenckstern, J. B. Varley, and T. D. Veal, ACS Appl. Mater. Interf., 13, 2807 (2021).
- 22. Y. Oshima, K. Kawara, T. Shinohe, T. Hitora, M. Kasu, and S. Fujita, APL Mater., 7. 022503 (2019).
- 23. M. Higashiwaki and S. Fujita (ed.), Gallium Oxide, Materials, Properties and Devices (Springer, Berlin) (2020).
- 24. H. Sun, K.-H. Li, C. G. T. Castanedo, S. Okur, G. S. Tompa, T. Salagaj, S. Lopatin, A. Genovese, and X. Li, Cryst. Growth Des., 18, 2370 (2018).
- 25. B. W. Krueger, C. S. Dandeneau, E. M. Nelson, S. T. Dunham, F. S. Ohuchi, and M. A. Olmstead, J. Am. Ceram. Soc., 99, 2467 (2016).
- 26. Y. W. Zhang et al., Appl. Phys. Lett., 112, 173502 (2018).
- 27. F. B. Zhang, K. Saito, T. Tanaka, M. Nishio, M. Arita, and Q. X. Guo, *Appl. Phys.* Lett., 105, 162107 (2014).
- 28. T. Uchida, R. Jinno, S. Takemoto, K. Kaneko, and S. Fujita, Jpn. J. Appl. Phys., 57, 040314 (2018).
- 29. Q. Feng, X. Li, G. Han, L. Huang, F. Li, W. Tang, J. Zhang, and Y. Haoet, Opt. Mater. Express, 7, 1240 (2017).
- 30. C. Kranert, M. Jenderka, J. Lenzner, M. Lorenz, H. von Wenckstern, R. Schmidt-Grund, and M. Grundmann, J. Appl. Phys., 117, 125703 (2015).
- 31. B. Mazumder, J. Sarker, Y. Zhang, J. M. Johnson, M. Zhu, S. Rajan, and J. Hwang, Appl. Phys. Lett., 115, 132105 (2019).
- 32. B. Mazumdera and J. Sarker, J. Mater. Res., 36, 52 (2020).
- 33. S.-H. Yuan, C.-C. Wang, S.-Y. Huang, and D.-S. Wuu, IEEE Electr. Dev. Lett., 39, 220 (2018).
- 34. G. T. Dang, T. Yasuoka, Y. Tagashira, T. Tadokoro, W. Theiss, and T. Kawaharamura, Appl. Phys. Lett., 113, 062102 (2018).
- 35. E. Ahmadi, O. S. Koksaldi, X. Zheng, T. Mates, Y. Oshima, U. K. Mishra, and J. S. Speck, Appl. Phys. Express, 10, 071101 (2017).
- 36. S. Krishnamoorthy et al., Appl. Phys. Lett., 111, 023502 (2017).
- 37. Y. Zhang, C. Joishi, Z. Xia, M. Brenner, S. Lodha, and S. Rajan, Appl. Phys. Lett., 112, 233503 (2018).
- 38. J. B. Varley, A. Perron, V. Lordi, D. Wickramaratne, and J. L. Lyons, Appl. Phys. Lett., 116, 172104 (2020).
- 39. A. F. M. Anhar Uddin Bhuiyan, Z. Feng, J. M. Johnson, Z. Chen, H. L. Huang, J. Hwang, and H. Zhao, Appl. Phys. Lett., 115, 120602 (2019).
- 40. R. Wakabayashi, K. Yoshimatsu, M. Hattori, J.-S. Lee, O. Sakata, and A. Ohtomo, Cryst. Growth Des., 21, 2844 (2021).
- 41. S. W. Kaun, F. Wu, and J. S. Speck, J. Vac. Sci. Technol. A, 33, 041508 (2015).
- 42. R. Miller, F. Alema, and A. Osinsky, IEEE Trans. Semicond. Manuf., 31, 467 (2018).
- 43. A. F. M. A. U. Bhuiyan, Z. Feng, J. M. Johnson, H. L. Huang, J. Sarker, M. Zhu, M. R. Karim, B. Mazumder, J. Hwang, and H. Zhao, *APL Mater.*, **8**, 031104 (2020). 44. J. Sarker, S. Broderick, A. F. M. A. U. Bhuiyan, Z. Feng, H. Zhao, and
- B. Mazumder, *Appl. Phys. Lett.*, **116**, 152101 (2020).
  45. H. Peelaers, J. B. Varley, J. S. Speck, and C. G. Van de Walle, *Appl.Phys. Lett.*,
- 112, 242101 (2018).
- 46. H. Kim, H. Ko, Y.-C. Chung, and S. B. Cho, J. Eur. Ceram. Soc., 41, 611 (2020).
- 47. T. Wang, W. Li, C. Ni, and A. Janotti, Phys. Rev. Appl., 10, 011003 (2018).
- 48. M. Hilfiker, U. Kilic, A. Mock, V. Darakchieva, S. Knight, R. Korlacki, A. Mauze, Y. Zhang, J. Speck, and M. Schubert, Appl. Phys.Lett., 114, 231901 (2019).
- 49. C. Fares, Z. Islam, A. Haque, M. Kneiß, H von Wenckstern, M. Grundmann, M. Tadjer, F. Ren, and S. J. Pearton, ECS J. Solid State Sci. Technol., 8, P751 (2019).
- 50. C. Fares, M. Kneiß, H von Wenckstern, M. Tadjer, F. Ren, E. Lambers, M. Grundmann, and S. J. Pearton, ECS J. Solid State Sci. Technol., 8, O P351 (2019).
- 51. H. von Wenckstern, M. Kneiß, A. Hassa, P. Storm, D. Splith, and M. Grundmann, Physica Status Solidi (B) Basic Research, 257, 1900626 (2019).
- 52. H. von Wenckstern, Z. Zhang, F. Schmidt, J. Lenzner, H. Hochmuth, and M. Grundmann, Cryst Eng Comm, 15, 10020 (2013).
- 53. K. B. Jinesh, J. L. van Hemmen, M. C. M. van de Sanden, F. Roozeboom, J. H. Klootwijk, W. F. A. Besling, and W. M. M. Kessels, J. Electrochem. Soc., 158, G21 (2011).
- 54. H. N. Masten, J. D. Phillips, and R. L. Peterson, IEEE Trans Electron Dev., 66, 2489 (2019).
- 55. D. C. Hays, B. P. Gila, S. J. Pearton, and F. Ren, Appl. Phys. Rev., 4, 021301 (2017).
- 56. J. B. Varley, J. Mater. Res. (2021), in press.