

Lattice-matched $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3/\beta\text{-Ga}_2\text{O}_3$ heterostructures with widely tunable bandgap

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Abstract

Bandgap engineering of $\beta\text{-Ga}_2\text{O}_3$ is essential for advancing its electronic and optoelectronic applications. However, the growth of high-quality heteroepitaxial structures is often hampered by large lattice mismatches. In this study, we design $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ quaternary alloys in the form of both polycrystalline powders and heteroepitaxial films and demonstrate their lattice matching to $\beta\text{-Ga}_2\text{O}_3$. Powder x-ray diffraction (XRD) measurements reveal that the lattice parameters of monoclinic $(\text{Al}_x\text{Sc}_y\text{Ga}_{0.8})_2\text{O}_3$ match those of $\beta\text{-Ga}_2\text{O}_3$ when $x/y = 1.5\text{--}3.2$. $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ thin films are grown on $\beta\text{-Ga}_2\text{O}_3$ (100) substrates by pulsed-laser deposition by varying the composition ratio x/y and the total substitutional fraction $x + y$. Nearly perfect lattice-matched epilayers with uniform composition and crystal structure are obtained up to $x + y \sim 0.6$, as confirmed by XRD and scanning transmission electron microscopy. Electron energy loss spectroscopy reveals a tunable bandgap from 4.5 to 5.8 eV. These results demonstrate that $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3/\beta\text{-Ga}_2\text{O}_3$ heterostructures are promising platforms for ultrawide-bandgap semiconductor devices.

Main text

Monoclinic β -Ga₂O₃ is an ultrawide-bandgap semiconductor that has received significant research interest over the past decades. Its large bandgap (E_g) of 4.4–5.0 eV,^{1,2} tunable n -type doping concentration ranging from 10^{15} to 10^{20} cm⁻³,^{3,4} and large melt-grown single-crystal wafers⁵ make it a promising candidate for high-voltage power devices.⁶ In addition, β -Ga₂O₃ is also promising in optoelectronic applications such as solar-blind UV detection⁷ and UV-transparent conductive electrodes⁸ owing to its high transparency down to a wavelength of 280 nm.

Bandgap engineering expands the design space of such devices. For example, the bandgap widening of β -Ga₂O₃ can enhance its durability under high electric fields and tune its spectral response in the UV-C range. Furthermore, heteroepitaxial structures with different E_g enable the control of charge-carrier profiles in electronic and photonic devices, such as modulation-doped field-effect transistors (MODFET), resonant tunneling diodes (RTD), and quantum-well infrared photodetectors (QWIP), which are similar to the applications of III-V semiconductors.

In analogy with cubic Al_xGa_{1-x}As and hexagonal Al_xGa_{1-x}N, the heteroepitaxy of monoclinic (Al_xGa_{1-x})₂O₃ has been investigated since the early stages of β -Ga₂O₃ research,⁹ although its parent oxides exhibit different crystal structures. The (010)-oriented (Al_xGa_{1-x})₂O₃/ β -Ga₂O₃ heterojunctions with x up to ~ 0.2 were implemented in MODFET¹⁰ and RTD.¹¹ However, the lattice mismatches between (Al_xGa_{1-x})₂O₃ and β -Ga₂O₃ amount to 3.2–4.4% when $x = 1$.^{12,13} As a result, heteroepitaxy of (Al_xGa_{1-x})₂O₃ on β -Ga₂O₃ has faced some constraints depending on the crystal orientation. On the β -Ga₂O₃ (010) substrate, phase transformation to the spinel-like γ -phase limits x to less than 0.27,¹⁴ and crack formation occurs in epilayers thicker than 100 nm when $x > 0.1$.¹⁵ On the β -Ga₂O₃ (100) substrate, strain relaxation proceeds in 50-nm-thick epilayers with $x = 0.28$,¹⁶ while 5-nm-thick epilayers (Al_xGa_{1-x})₂O₃ can be epitaxially stabilized in the entire composition range.¹⁷ On the β -Ga₂O₃ (001) substrate, inhomogeneous cation distribution occurs in epilayers with an average $x = 0.14$.¹⁸ Substantially, the tunable E_g increment without introducing lattice disorders is approximately 0.4 eV¹⁹,

48 which is recognized as the upper limit of the design space with $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$.

49 A lattice-matched quaternary alloy is an effective approach to address these issues.
50 Considering the ionic radius, $(\text{Al,Ga,In})_2\text{O}_3$ and $(\text{Al,Sc,Ga})_2\text{O}_3$ alloys can maintain lattice parameters
51 close to those of $\beta\text{-Ga}_2\text{O}_3$ under specific Al:In or Al:Sc ratios, since the ionic radii of Al^{3+} (Sc^{3+} and
52 In^{3+}) are smaller (larger) than those of Ga^{3+} .²⁰ Recently, $(\text{Al,Ga,In})_2\text{O}_3$ quaternary alloys were
53 investigated both theoretically²¹ and experimentally.^{22,23} However, the lattice-matched
54 $(\text{Al}_{0.17}\text{Ga}_{0.76}\text{In}_{0.07})_2\text{O}_3/\beta\text{-Ga}_2\text{O}_3$ (010) heterostructure exhibited inhomogeneous In incorporation and
55 local γ -phase inclusion. On the other hand, $(\text{Al,Sc,Ga})_2\text{O}_3$ alloys gain an advantage over $(\text{Al,Ga,In})_2\text{O}_3$
56 since both Al- and Sc-substitutions contribute to the increase in E_g ,^{16,19,24,25} while co-substitution by
57 Al and In partially cancels E_g variation.^{21–23} Additionally, Sc incorporation appears easier than In due
58 to the absence of competitive suboxide reactions as observed for In.^{26,27} We thus examine the variation
59 in lattice parameters in polycrystalline $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ powder samples to identify the lattice-
60 matching composition ratio x/y and fabricate (100)-oriented $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers with the
61 appropriate ratios to demonstrate lattice-matched heterostructures with large tunability of E_g .

62

63 Polycrystalline $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ powders (Toshiba Manufacturing, 3N purity) were
64 prepared to investigate their crystal structures. For simplicity, the substitutional molar fraction was
65 fixed at $x + y = 0.2$, while the $x:y$ ratio was varied as 20:0, 15:5, 10:10, 5:15, and 0:20.

66 $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers were grown on $\beta\text{-Ga}_2\text{O}_3$ (100) substrates (Novel Crystal
67 Technology) by oxygen-radical-assisted pulsed-laser deposition (PLD).²⁸ For all runs, the substrate
68 temperature, fluence, and frequency of the KrF laser pulses were kept at 750 °C, 0.4 J cm^{-2} , and 10
69 Hz, respectively. Oxygen radicals were supplied from an RF plasma cell operating at 200 W with an
70 O_2 flow rate of 1.00 sccm, corresponding to a constant pressure of 1.5×10^{-4} Torr in the PLD chamber.
71 The epilayers were grown either by ablating a single target or by alternately ablating two different

72 targets, which were ceramics or single crystals. The layer thickness was regulated using the intensity
73 oscillation period of the reflection high-energy electron diffraction (RHEED) spots. Further details of
74 the deposition procedure are provided in the supplementary material.

75 To investigate structural properties, x-ray diffraction (XRD) measurements were carried out
76 on the polycrystalline powder and the epilayer samples with Cu K α and Cu K α_1 radiation, respectively.
77 The composition of the epilayers was analyzed by Auger electron spectroscopy (AES) using scanning
78 electron microscopy (JAMP-9500F, JEOL). (Al $_x$ Ga $_{1-x}$) $_2$ O $_3$ and (Sc $_y$ Ga $_{1-y}$) $_2$ O $_3$ ceramics were used as
79 standards for calibrating the composition. To investigate microstructures and composition of the
80 epilayers, cross-sectional scanning transmission electron microscopy (STEM) observations were
81 performed using an aberration-corrected STEM (Spectra Ultra, Thermo Fisher Scientific) equipped
82 with an energy-dispersive x-ray spectroscopy (EDS) detector. A cross-sectional thin-foiled specimen
83 with a thickness of approximately 100 nm was prepared by focused ion beam process using NB5000
84 (Hitachi High-Tech) dual-beam system, followed by Ar ion milling using a PIPS II (Gatan). The E_g
85 values were estimated by reflection electron energy loss spectroscopy (REELS) in the AES apparatus.

86
87 Powder-XRD measurement confirmed that all the (Al $_x$ Sc $_y$ Ga $_{0.8}$) $_2$ O $_3$ samples maintained a
88 pure monoclinic phase (see Fig. S1 in the supplementary material). Table I summarizes their lattice
89 parameters. As shown in Fig. 1(a), the lattice parameters a , b , and c (β) increased (decreased) as the
90 Sc content increased. From linear regression, the x - and y -dependencies of the lattice parameters'
91 variations with respect to β -Ga $_2$ O $_3$ were formulated to be $\Delta a = -0.47x + 1.1y$ (Å), $\Delta b = -0.13x + 0.26y$
92 (Å), $\Delta c = -0.17x + 0.26y$ (Å), and $\Delta\beta = 0.71x - 2.3y$ (deg). Therefore, the individual lattice parameters
93 match those of β -Ga $_2$ O $_3$ when x/y is between 1.5 and 3.2. This range of x/y values is reasonable, as it
94 centers on a specific ratio of $x/y = 2.4$, where the average cation radius of Al $^{3+}$ and Sc $^{3+}$ is equal to that
95 of Ga $^{3+}$.²⁰ Although exact lattice matching is unattainable in monoclinic β -Ga $_2$ O $_3$ due to four lattice
96 parameters varying independently, a nearly perfect lattice-matched (Al $_x$ Sc $_y$ Ga $_{1-x-y}$) $_2$ O $_3$ / β -Ga $_2$ O $_3$

heterostructure is feasible within this compositional range.

Having established the lattice-matching x/y range, ~60-nm-thick $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers were grown on a $5\times 10\text{ mm}^2$ substrate using a combinatorial approach. A continuous composition gradient was created such that the x/y ratio varied along the 10-mm-length direction, while the total substitutional fraction $x + y$ was fixed to 0.45 ± 0.02 . This total amount of substitution was beyond the composition thresholds for strain relaxation in ternary $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ and $(\text{Sc}_y\text{Ga}_{1-y})_2\text{O}_3$ epilayers having similar thicknesses.^{16,24} Figure 1 (b) shows reciprocal space maps (RSMs) around 710 reflections from the selected sample area, corresponding to epilayers with $(x, y, x/y) = (0.23, 0.21, 1.1)$, $(0.30, 0.15, 2.0)$, $(0.35, 0.12, 2.9)$, and $(0.36, 0.07, 5.1)$, respectively. An increase in Q_z of the reflections from the epilayers reflects the shrinkage of the out-of-plane lattice spacing with increasing (decreasing) Al (Sc) fraction, as indicated by the a/a_0 of the powder samples. All of the primary reflections from the epilayers were found at the same Q_x value as the substrate, regardless of the composition, indicating that the epilayers were coherently strained along the [010] direction. In addition, the sample with the highest Sc content exhibited a diffuse component, and its centroid shifted toward a smaller Q_x compared to that of the substrate. These features suggest not only partial relaxation of the compressive strain arising from out of the lattice-matching x/y range but also the reduction of the lateral coherence length²⁹ along the [010] direction. Similar features were observed in the sample with the highest Al content. However, the direction of the centroid shift was opposite to that of the Sc-rich case due to the relaxation of opposite strain, i.e., tensile strain. In contrast, the other two samples, having $x/y = 2.0$ and 2.9 , showed only sharp reflection spots, indicating an absence of strain relaxation owing to the compositions within the lattice-matching range ($1.5 \leq x/y \leq 3.2$).

Next, a series of nearly lattice-matched ~20-nm-thick $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers was grown using single ceramic targets with fixed $x/y = 2$ and various $x + y$ values of 0.2, 0.4, and 0.6. The composition parameters $(x, y, x/y)$ of the epilayers were $(0.21, 0.07, 3.0)$, $(0.31, 0.11, 2.8)$, and $(0.48, 0.15, 3.2)$, respectively. A slight deviation from the target compositions is likely due to differences in

the laser ablation efficiency and/or preferential scattering of constituent cations.³⁰ Figure 2 shows out-of-plane XRD profiles around the substrates' 400 reflection for the epilayers. The 400 reflections of the epilayers overlapped with those of the substrates at $2\theta \sim 30^\circ$. In contrast, oscillatory fringes appeared in their tails, known as Laue oscillations, indicating structural homogeneity and smooth surface/interfaces³¹ even at a very high substitutional fraction of $x + y = 0.63$.

Cross-sectional STEM observations were performed along the [010] zone axis of an $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayer. A high-angle annular dark-field (HAADF)-STEM image and the corresponding EDS chemical maps, presented in Figs. 3(a–d), determined the layer thickness and average composition parameters to be 73 nm and $(x, y, x/y) = (0.38, 0.18, 2.1)$, respectively. As the intensity in a HAADF-STEM image correlates with the atomic number of the constituent elements, the homogeneous, darker contrast of the epilayer informs a higher $x + y$ value without noticeable segregation of Al and Sc at the observed scale. The homogeneous cation distribution was also confirmed by the EDS maps of Al, Sc, and Ga, as shown in Figs. 3(b–d), respectively.

Figure 3(e) shows a HAADF-STEM image near the substrate/epilayer interface, revealing an atomically flat and compositionally abrupt interface. In the epilayer, however, the cation arrangement is partially altered, which is attributed to twinning along the [001] direction, as illustrated in Fig. 3(f). Such twinning has been reported during the epitaxial growth of (100)-oriented $\beta\text{-Ga}_2\text{O}_3$ and $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$.^{32,33} The observed stacking sequence in Fig. 3(f) can be described as follows: (i) stacking of one and a half unit cells along the [100] direction, (ii) stacking of a half twinned unit cell with a displacement of $c/2$, forming a twin boundary on the (100)A plane [$\text{TB}_{(100)\text{A}}$],³⁴ (iii) stacking of one twinned unit cell that coalesces with untwinned regions, forming a second $\text{TB}_{(100)\text{A}}$ and twin boundaries on the $(\bar{1}02)$ plane [$\text{TB}_{(-102)}$]³⁴ arranged with a spacing of c , and (iv) stacking of two or more unit cells on a third $\text{TB}_{(100)\text{A}}$. $\text{TB}_{(100)\text{A}}$ can be readily formed due to its low formation energy, whereas $\text{TB}_{(-102)}$ has a much higher formation energy,³⁴ which likely causes $\text{TB}_{(-102)}$ to terminate within one unit cell along the growth direction, being consistent with the STEM observations. Among

the observed TBs, $TB_{(-102)}$ is classified as an incoherent TB that deteriorates the electrical transport properties.³² Generally, growth on miscut substrates is effective for eliminating stacking faults and/or TBs. Indeed, the use of a 6°-off (100) substrate along the $[00\bar{1}]$ direction has enabled TB-free step flow growth in homoepitaxy.^{35,36}

Figure 4 (a) shows the room-temperature energy loss spectra measured for ~20-nm-thick $(Al_xSc_yGa_{1-x-y})_2O_3$ epilayers (which are identical to those shown in Fig. 2) together with a reference homoepitaxial β -Ga₂O₃ (100) film. In the spectra, the energy of the intensity onset (E_{onset}) shifts to the higher loss-energy side, and the intensity above 8 eV increases with increasing $x + y$ value. The former reflects an increase in E_g , while the latter is attributed to an increase in the unoccupied Sc 3d density of states. The E_{onset} values were determined by extrapolating the linear portions of the slopes to the background level. Since the E_{onset} value of β -Ga₂O₃ (4.5 eV) agrees with the lowest direct optical bandgap (4.48 eV) of a bulk single crystal,¹ it can be regarded as a proxy for E_g . Figure 4(b) compares E_{onset} values of quaternary $(Al_xSc_yGa_{1-x-y})_2O_3$ epilayers as a function of out-of-plane lattice spacing d_{100} with those of ~20-nm-thick ternary $(Al_xGa_{1-x})_2O_3$ and $(Sc_yGa_{1-y})_2O_3$ epilayers. The d_{100} values for the ternary epilayers were determined from the 2θ peak positions of the 400 reflections in the XRD profiles, whereas for the quaternary epilayers, the d_{100} value of the β -Ga₂O₃ substrate was used. An attainable E_{onset} value of the lattice-matched $(Al_xSc_yGa_{1-x-y})_2O_3$ epilayers was up to 5.8 eV, whereas that of the coherently strained $(Al_xGa_{1-x})_2O_3$ and $(Sc_yGa_{1-y})_2O_3$ epilayers was limited to 5.3 eV and 5.2 eV, respectively, due to the strain relaxation.

The E_{onset} range between β -Ga₂O₃ and $(Al_xSc_yGa_{1-x-y})_2O_3$ spans 4.5–5.8 eV, which is larger in both absolute energy and energy width than those reported for conventional lattice-matched heterojunctions [Fig. 4(c)]. Furthermore, since the valence band maximum of β -Ga₂O₃ is primarily composed of O 2p states, a small valence-band offset, and consequently, a large conduction-band offset (ΔE_c) are anticipated at the $(Al_xSc_yGa_{1-x-y})_2O_3/\beta$ -Ga₂O₃ heterojunction. These features enable the formation of lattice-matched ultrawide-bandgap heterostructures with high structural quality and wide

172 tunability of E_g and ΔE_c . Such wide tunability is particularly advantageous for high-temperature and
173 high-voltage device operation of MODFETs and RTDs, where both high breakdown field and carrier
174 confinement are required.

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176 This study presents structural and electronic properties of quaternary $(Al_xSc_yGa_{1-x-y})_2O_3$
177 lattice matched to β - Ga_2O_3 . Monoclinic $(Al_xSc_yGa_{0.8})_2O_3$ polycrystals were successfully synthesized,
178 and their lattice parameters varied by x and y , reflecting the differences in ionic radii. The four
179 independent lattice parameters were found to match those of β - Ga_2O_3 in the x/y range of 1.5–3.2.
180 Epitaxial growth of $(Al_xSc_yGa_{1-x-y})_2O_3$ on β - Ga_2O_3 (100) substrates, as confirmed by RSM
181 measurements, yielded coherently strained epilayers with $x + y \sim 0.45$, exceeding the critical
182 composition achievable in ternary $(Al_xGa_{1-x})_2O_3$ and $(Sc_yGa_{1-y})_2O_3$ epilayers. Additionally, a high
183 substitutional fraction up to $x + y = 0.63$ was achieved in nearly lattice-matched $(Al_xSc_yGa_{1-x-y})_2O_3$
184 epilayers. STEM and EDS measurements of the $(Al_{0.38}Sc_{0.18}Ga_{0.44})_2O_3$ epilayer revealed its
185 homogeneous cation distribution and a compositionally abrupt substrate/epilayer interface. The
186 formation of twin boundaries was also observed due to the specific substrate orientation, which could
187 be eliminated by the use of miscut substrates. The REELS spectra of the quaternary epilayers
188 exhibited an increase in E_{onset} from 4.5 eV ($x = y = 0$) to 5.8 eV ($x = 0.48$, $y = 0.15$). Therefore, this
189 quaternary system uniquely realizes wide E_g tunability while maintaining near lattice-matching to β -
190 Ga_2O_3 , which is not achievable with ternary alloys. Furthermore, the large E_g and its wide tunability
191 are exceptional among lattice-matched heterojunctions. These capabilities open a pathway to β -
192 Ga_2O_3 -based bandgap-engineered ultrawide-bandgap power electronic and optoelectronic devices,
193 particularly those relying on high-quality heterojunctions.

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195 See the supplementary material for detailed descriptions of the epitaxial growth and XRD
196 patterns of the $(Al_xSc_yGa_{1-x-y})_2O_3$ powder samples available online.

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TABLE I. Lattice parameters of $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ powder samples calculated from powder XRD data.

x	y	a (Å)	b (Å)	c (Å)	β (deg)
0	0	12.22	3.04	5.81	103.84
0	0.20	12.44	3.09	5.86	103.39
0.05	0.15	12.37	3.07	5.84	103.55
0.10	0.10	12.29	3.05	5.82	103.68
0.15	0.05	12.21	3.03	5.80	103.84
0.20	0	12.13	3.01	5.78	103.99

211 **References**

- 212 ¹ T. Onuma, S. Saito, K. Sasaki, T. Masui, T. Yamaguchi, T. Honda, and M. Higashiwaki, *Jpn. J.*
213 *Appl. Phys.* **54**, 112601 (2015).
- 214 ² A. Mock, R. Korlacki, C. Briley, V. Darakchieva, B. Monemar, Y. Kumagai, K. Goto, M.
215 Higashiwaki, and M. Schubert, *Phys. Rev. B* **96**, 245205 (2017).
- 216 ³ C. Peterson, A. Bhattacharyya, K. Chanchaiworawit, R. Kahler, S. Roy, Y. Liu, S. Rebollo, A.
217 Kallistova, T.E. Mates, and S. Krishnamoorthy, *Appl. Phys. Lett.* **125**, 182103 (2024).
- 218 ⁴ H.M. Jeon, K.D. Leedy, D.C. Look, C.S. Chang, D.A. Muller, S.C. Badescu, V. Vasilyev, J.L.
219 Brown, A.J. Green, and K.D. Chabak, *APL Mater.* **9**, 101105 (2021).
- 220 ⁵ T. Igarashi, Y. Ueda, K. Koshi, R. Sakaguchi, S. Watanabe, S. Yamakoshi, and A. Kuramata, *Phys.*
221 *Status Solidi* **262**, 2400444 (2025).
- 222 ⁶ A.J. Green, J. Speck, G. Xing, P. Moens, F. Allerstam, K. Gumaelius, T. Neyer, A. Arias-Purdue,
223 V. Mehrotra, A. Kuramata, K. Sasaki, S. Watanabe, K. Koshi, J. Blevins, O. Bierwagen, S.
224 Krishnamoorthy, K. Leedy, A.R. Arehart, A.T. Neal, S. Mou, S.A. Ringel, A. Kumar, A. Sharma, K.
225 Ghosh, U. Singiseti, W. Li, K. Chabak, K. Liddy, A. Islam, S. Rajan, S. Graham, S. Choi, Z. Cheng,
226 and M. Higashiwaki, *APL Mater.* **10**, 029201 (2022).
- 227 ⁷ J. Xu, W. Zheng, and F. Huang, *J. Mater. Chem. C* **7**, 8753 (2019).
- 228 ⁸ J. Zhang, J. Willis, Z. Yang, X. Lian, W. Chen, L.-S. Wang, X. Xu, T.-L. Lee, L. Chen, D.O.
229 Scanlon, and K.H.L. Zhang, *Cell Reports Phys. Sci.* **3**, 100801 (2022).
- 230 ⁹ T. Oshima, T. Okuno, N. Arai, Y. Kobayashi, and S. Fujita, *Jpn. J. Appl. Phys.* **48**, 070202 (2009).
- 231 ¹⁰ Y. Zhang, A. Neal, Z. Xia, C. Joishi, J.M. Johnson, Y. Zheng, S. Bajaj, M. Brenner, D. Dorsey, K.
232 Chabak, G. Jessen, J. Hwang, S. Mou, J.P. Heremans, and S. Rajan, *Appl. Phys. Lett.* **112**, 173502
233 (2018).
- 234 ¹¹ H. Okumura, *Jpn. J. Appl. Phys.* **59**, 075503 (2020).
- 235 ¹² J. Åhman, G. Svensson, and J. Albertsson, *Acta Crystallogr. Sect. C Cryst. Struct. Commun.* **52**,
236 1336 (1996).
- 237 ¹³ Y.R. E. Husson, *Eur. J. Solid State Inorg. Chem.* **33**, 1223 (1996).
- 238 ¹⁴ A.F.M.A.U. Bhuiyan, Z. Feng, J.M. Johnson, H.-L. Huang, J. Sarker, M. Zhu, M.R. Karim, B.
239 Mazumder, J. Hwang, and H. Zhao, *APL Mater.* **8**, 031104 (2020).
- 240 ¹⁵ J.S. Lundh, K. Huynh, M. Liao, W. Olsen, K. Pan, K. Sasaki, K. Konishi, H.N. Masten, J.K. Hite,
241 M.A. Mastro, N.A. Mahadik, M. Goorsky, A. Kuramata, K.D. Hobart, T.J. Anderson, and M.J.
242 Tadjer, *Appl. Phys. Lett.* **123**, 222104 (2023).

243 ¹⁶ R. Wakabayashi, M. Hattori, K. Yoshimatsu, K. Horiba, H. Kumigashira, and A. Ohtomo, Appl.
 244 Phys. Lett. **112**, 232103 (2018).
 245 ¹⁷ R. Wakabayashi, K. Yoshimatsu, M. Hattori, J.-S. Lee, O. Sakata, and A. Ohtomo, Cryst. Growth
 246 Des. **21**, 2844 (2021).
 247 ¹⁸ A.F.M.A. Uddin Bhuiyan, L. Meng, H.L. Huang, J. Sarker, C. Chae, B. Mazumder, J. Hwang, and
 248 H. Zhao, APL Mater. **11**, 041112 (2023).
 249 ¹⁹ M. Hilfiker, U. Kilic, A. Mock, V. Darakchieva, S. Knight, R. Korlacki, A. Mauze, Y. Zhang, J.
 250 Speck, and M. Schubert, Appl. Phys. Lett. **114**, 231901 (2019).
 251 ²⁰ R.D. Shannon, Acta Crystallogr. Sect. A **32**, 751 (1976).
 252 ²¹ S. Seacat, J.L. Lyons, and H. Peelaers, Phys. Rev. Mater. **8**, 014601 (2024).
 253 ²² L. Vasylechko, V. Hreb, Y. Zhydashkevskyy, Y. Hizhnyi, A. Smaliuk, V. Stasiv, V. Stadnik, Y.
 254 Hirskeyi, H. Zhydashkevskya, V. Mykhaylyk, and A. Suchocki, Sci. Rep. **15**, 37128 (2025).
 255 ²³ S. Schaefer, M. Smeaton, K. Egbo, S. Hasan, W. Callahan, G. Teeter, A. Zakutayev, and M.B.
 256 Tellekamp, Appl. Phys. Lett. **125**, 172106 (2024).
 257 ²⁴ K. Koreishi, T. Soma, H. Kumigashira, and A. Ohtomo, Appl. Phys. Lett. **125**, 152101 (2024).
 258 ²⁵ Q. Wang, P. Huang, Q. Liu, Y. Li, Q. Qu, M. Li, K.P. Homewood, Y. Lu, and Y. He, J. Alloys
 259 Compd. **834**, 155036 (2020).
 260 ²⁶ P. Mazzolini, C. Wouters, M. Albrecht, A. Falkenstein, M. Martin, P. Vogt, and O. Bierwagen,
 261 ACS Appl. Mater. Interfaces **16**, 12793 (2024).
 262 ²⁷ S. Schaefer, D. Febba, K. Egbo, G. Teeter, A. Zakutayev, and B. Tellekamp, J. Mater. Chem. A
 263 **12**, 5508 (2024).
 264 ²⁸ R. Wakabayashi, T. Oshima, M. Hattori, K. Sasaki, T. Masui, A. Kuramata, S. Yamakoshi, K.
 265 Yoshimatsu, and A. Ohtomo, J. Cryst. Growth **424**, 77 (2015).
 266 ²⁹ R. Chierchia, T. Böttcher, H. Heinke, S. Einfeldt, S. Figge, and D. Hommel, J. Appl. Phys. **93**,
 267 8918 (2003).
 268 ³⁰ S. Wicklein, A. Sambri, S. Amoruso, X. Wang, R. Bruzzese, A. Koehl, and R. Dittmann, Appl.
 269 Phys. Lett. **101**, 131601 (2012).
 270 ³¹ A.M. Miller, M. Lemon, M.A. Choffel, S.R. Rich, F. Harvel, and D.C. Johnson, Zeitschrift Für
 271 Naturforsch. B **77**, 313 (2022).
 272 ³² A. Fiedler, R. Schewski, M. Baldini, Z. Galazka, G. Wagner, M. Albrecht, and K. Irmscher, J.
 273 Appl. Phys. **122**, 165701 (2017).
 274 ³³ A.F.M.A.U. Bhuiyan, L. Meng, H.L. Huang, C. Chae, J. Hwang, and H. Zhao, Phys. Status Solidi
 275 RRL **17**, 2300224 (2023).

276 ³⁴ M. Wang, S. Mu, J.S. Speck, and C.G. Van de Walle, Adv. Mater. Interfaces **2023**, 2300318
277 (2023).

278 ³⁵ R. Schewski, M. Baldini, K. Irmscher, A. Fiedler, T. Markurt, B. Neuschulz, T. Remmele, T.
279 Schulz, G. Wagner, Z. Galazka, and M. Albrecht, J. Appl. Phys. **120**, 225308 (2016).

280 ³⁶ P. Mazzolini, A. Falkenstein, Z. Galazka, M. Martin, and O. Bierwagen, Appl. Phys. Lett. **117**,
281 222105 (2020).

282 ³⁷ K. Momma and F. Izumi, J. Appl. Crystallogr. **44**, 1272 (2011).

283 ³⁸ I. Vurgaftman, J.R. Meyer, and L.R. Ram-Mohan, J. Appl. Phys. **89**, 5815 (2001).

284 ³⁹ S.N. Alam, V.Z. Zubialeovich, B. Ghafary, and P.J. Parbrook, Sci. Rep. **10**, 16205 (2020).
285
286

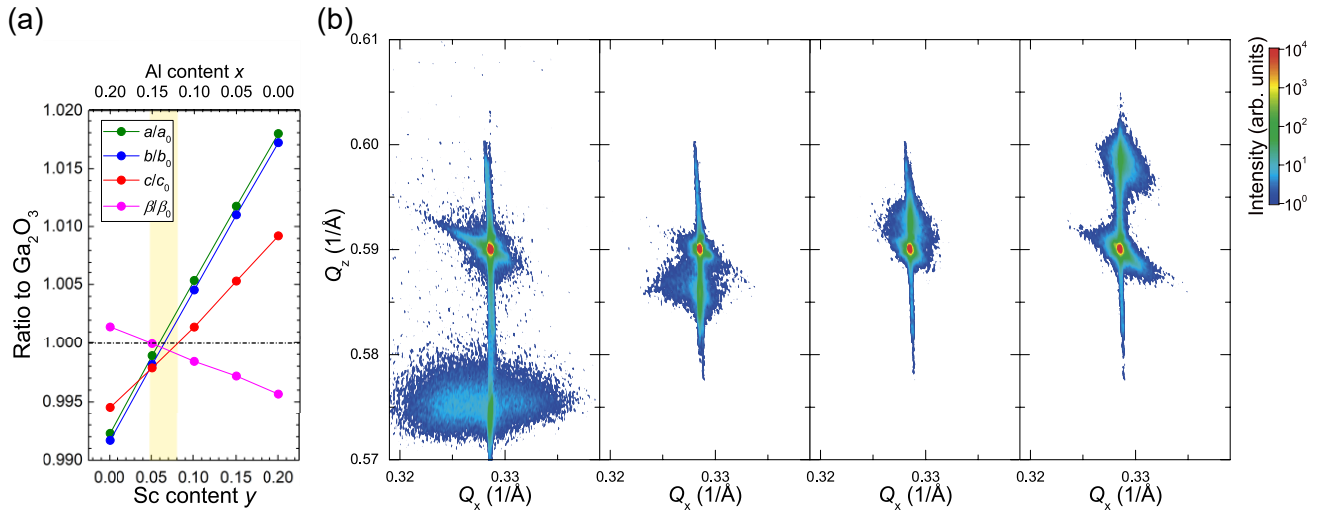


FIG. 1. (a) Lattice parameters of $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ powder samples having $x + y = 0.2$ with respect to those of $\beta\text{-Ga}_2\text{O}_3$. Individual lattice parameters match those of $\beta\text{-Ga}_2\text{O}_3$ within the highlighted region. (b) Reciprocal space maps around 710 reflections for $\sim 60\text{-nm}$ -thick $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers having $x + y \sim 0.45$ grown on $\beta\text{-Ga}_2\text{O}_3$ (100) substrates.

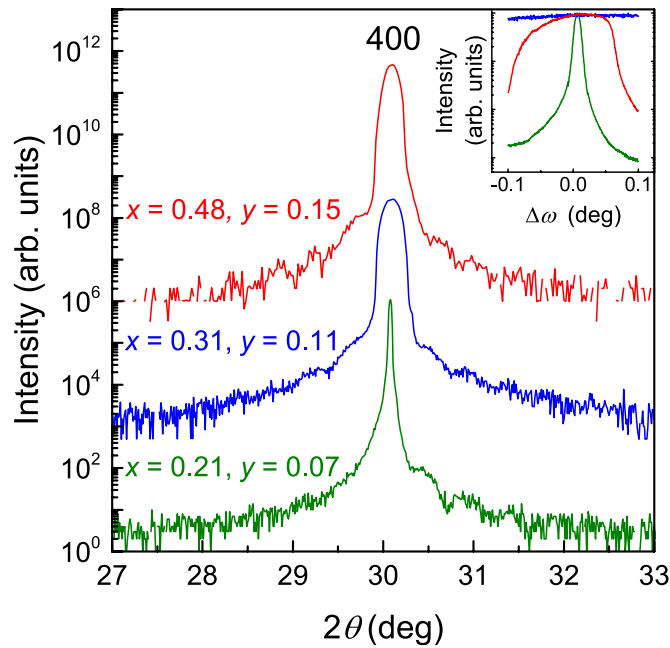


FIG. 2. Out-of-plane XRD profiles around substrates' 400 reflections for $\sim 20\text{-nm}$ -thick $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers. The inset shows ω scans of substrates' 400 reflections. Particularly broad peak shapes that are observed in the upper (red) and middle (blue) profiles stem from substrate bending confirmed in the corresponding ω scans. The substrate bending is sometimes initiated by the cutting process and/or clamping during the growth.

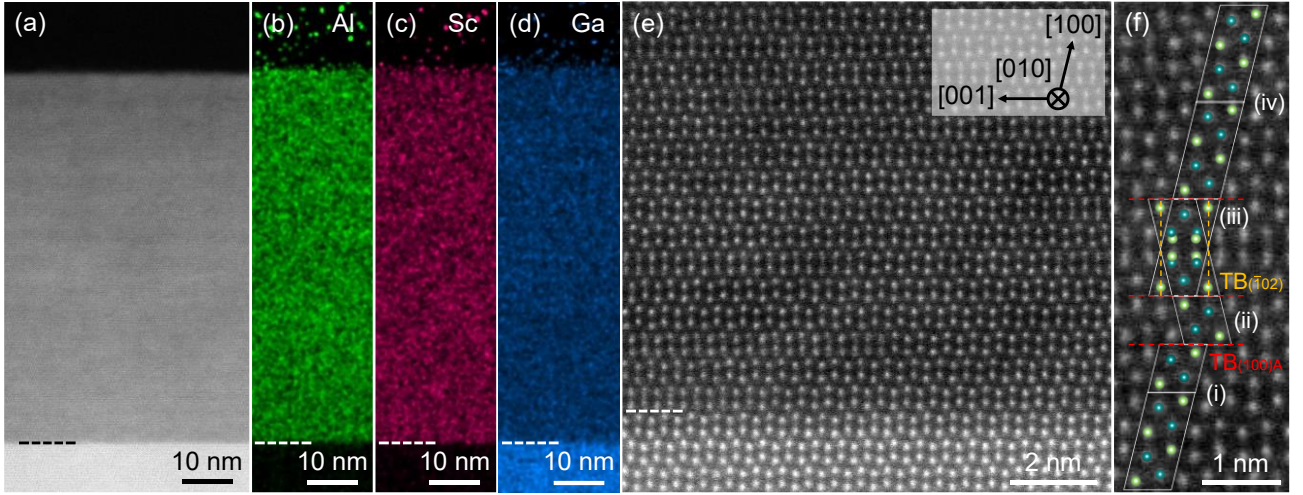


FIG. 3. Cross-sectional HAADF-STEM images and EDS maps of 73-nm-thick $(\text{Al}_{0.38}\text{Sc}_{0.18}\text{Ga}_{0.44})_2\text{O}_3$ epilayer. The solid lines in (a–e) indicate interfaces between the epilayer and $\beta\text{-Ga}_2\text{O}_3$ substrate. The atomic model in (f) shows one and a half cells of twinned and untwinned $\beta\text{-Ga}_2\text{O}_3$, where overlaid light- and deep-green balls represent cations at the oxygen tetrahedral and octahedral sites, respectively. The models were generated by VESTA.³⁷ These data were acquired at an acceleration voltage of 200 kV.

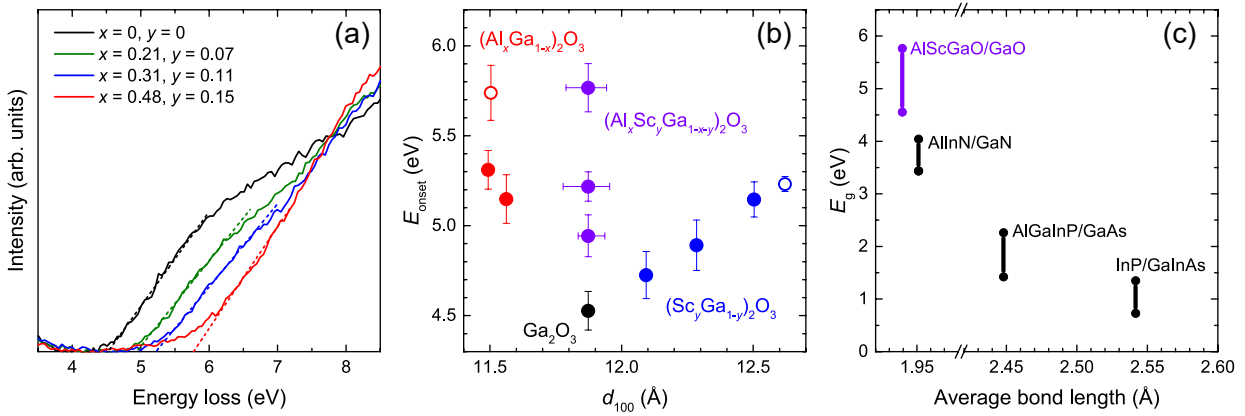


FIG. 4. (a) Energy loss spectra near the onset of loss peaks measured for ~ 20 -nm-thick $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ epilayers. The intercepts of the dashed lines correspond to E_{onset} . (b) E_{onset} as a function of out-of-plane lattice spacing d_{100} . Error bars along the x-axis for $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ and the y-axis correspond to the full width at half-maximum of substrates' 400 reflection in XRD and standard deviations of the intercept by linear fits in (a), respectively. The solid and open symbols represent coherently-strained and partially-relaxed epilayers, respectively. (c) E_g values vs. average bond length of selected semiconductors that can construct lattice-matched heterojunctions.^{38,39} Subscripts denoting the chemical composition are omitted. In AlScGaO/GaO , E_g is replaced with E_{onset} .