

β -Ga₂O₃-based heterojunctions: Effects of growth orientation and alloying on electronic properties

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We investigate the effects of alloying and growth orientation on the electronic properties of the ultrawide bandgap semiconductor β -Ga₂O₃ and pseudomorphic (Al_xGa_{1-x})₂O₃ alloy heterojunctions. Band offsets are computed from first principles using density functional theory with the Heyd-Scuseria-Ernzerhof hybrid functional for different aluminum concentrations and four growth orientations, namely, (100)B, (010), (001)B, and (201). Significant variations are found and ascribed to the strained pseudomorphic alloys. The values of the band offsets are fed into technology computer-aided design (TCAD) models of Schottky barrier diodes (SBD). *I-V* and *C-V* characteristics from the TCAD models show reasonable agreement with recent experimental measurements in the forward bias region. Discrepancies in the negative bias region are expected due to the ideality of the Schottky junctions considered in this study. Our findings underscore the critical role of growth orientation and strain in the accurate modeling of β -Ga₂O₃-based SBD.

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I. INTRODUCTION

Ga₂O₃, with its wide bandgap of approximately 4.7 eV, has garnered significant attention as a promising material for various applications, including MOSFETs, UV solar-blind photodetectors, and Schottky barrier diodes (SBDs) [1,2]. Al₂O₃ is frequently considered a gate oxide in Ga₂O₃-based heterojunctions [3–8]. Differences in the crystal structures of Ga₂O₃ and Al₂O₃ can lead to lattice mismatches in heterojunctions. Cracking may occur for film thicknesses exceeding a critical value [7,9]. To mitigate these mismatches—and also to engineer electronic properties such as the bandgap—alloying Ga₂O₃ with aluminum (Al) (Al_xGa_{1-x})₂O₃ is widely employed [4–8,10–12]. In applications such as SBD and high-electron-mobility transistors, band offset is crucial in developing semiconductor devices. The reported values of CBO can vary significantly, by up to 1.5 eV [5,8,13–22]. Experimental studies, such as those using capacitance-voltage (*C-V*) profiling and x-ray photoelectron spectroscopy (XPS), exhibit strong variation,

which also depends on the methods used for deposition (e.g., sputtering-prepared films, pulsed laser deposition, and atomic layer deposition) and the characterization techniques employed [19–22]. Theoretical studies, typically based on atomistic modeling, provide subresolution information regarding the large anisotropy of these alloys, where the monoclinic phase's low symmetry results in varying surface growth orientations. Most of the atomistic modeling reported in the literature considered isolated surfaces, i.e., offset computed using the electron affinity rule [5,8,16,17]. Although these models included surface relaxation, only a few studies considered proper interfaces to model these heterojunctions [13–15,18]. We note here that, while most of these reports investigate how the band offset and electron affinity are affected by both composition and structure, in nearly all cases, only the natural band offset is considered. In these cases, the effect of strain, as well as the chemical bonding at the interfaces, is neglected, which can lead to inaccurate predictions of both the magnitude and the nature of the band offset [13,18].

Here, using density functional theory (DFT) at the hybrid functional level, we investigate the β -Ga₂O₃/(Al_xGa_{1-x})₂O₃ interface. The accurate prediction of the band offset and its dependence on the crystallographic orientation within the material are crucial to improve the overall performance of devices. The monoclinic phase of Ga₂O₃ is considered as the substrate, assuming that the

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grown film has the same structure and orientation as the substrate, i.e., pseudomorphic growth. The calculated band offsets and affinities are integrated into technology computer-aided design (TCAD) models of SBD to study their electrical characteristics. In the following, we initially provide a detailed description of the modeling methodology, followed by the presentation of our findings regarding the electronic parameters of substrates and films, considering strain effects. Then, we explore the band offsets of pseudomorphically grown films $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$, exploring their significance for device design. We found that orientation and strain have a critical role in the accurate modeling of these heterojunctions and thus in optimizing wide-bandgap devices for power electronics.

II. COMPUTATIONAL DETAILS

The electronic structure was calculated using DFT with the Vienna *ab initio* simulation package [23–25]. For structural properties, full optimization, the strongly constrained and appropriately normed (SCAN) [26] functional was initially used. Additionally, for atomic relaxation and electronic properties, the Heyd-Scuseria-Ernzerhof (HSE06) [27,28] functional was used with a mixing parameter of 0.32. A gamma sampling scheme for the Brillouin zone was used, with a 0.03 (0.025) $1/\text{\AA}$ separation for structural (electronic) calculations and 600 eV as the plane wave cutoff energy. The projected augmented wave pseudopotential [29] was employed with the following valence configurations: $d^{10}s^2p^1$ for gallium (Ga), s^2p^1 for aluminum, and s^2p^4 for oxygen.

The structures were relaxed until the maximum force components reached a value less than 0.01 eV/ $\text{\AA}$, with an energy convergence threshold set to 10^{-6} eV for all supercell calculations. Both θ - Al_2O_3 and β - Ga_2O_3 crystallize in the monoclinic space group C^2/m , with 20-atom conventional unit cells [30]. In both compounds, trivalent Al or Ga cations occupy octahedral and tetrahedral sites. The interfaces were constructed using a periodic supercell along the out-of-plane direction, with no vacuum. We refer to this construction as the superlattice structures. The in-plane lattice constants of the films are fixed to those of the substrate.

Different orientations and the most stable terminations were considered, namely, (100), (010), (001), and $(\bar{2}01)$. In the cases of (100) and (001) orientations, there are two possible terminations. The most stable are (100)B and (001)B. (Superlattice structures are shown in Supplemental Material [33].) Only symmetric and nonpolar terminations were considered. The thickness of each layer of the superlattice structures (film: Al_2O_3 and substrate: Ga_2O_3) was set to ensure that the electron density in the middle of the layer is the same as in the bulk reference.

The average electrostatic (i.e., Hartree plus external) potential is used as a reference for bulk calculation and to

compute the potential lineup between both sides of the interface. The valence band offset (VBO) was computed as

$$\text{VBO}(A/B) = E_{\text{VBM,Ref}}^B - E_{\text{VBM,Ref}}^A + \Delta\bar{V}^{AB}, \quad (1)$$

where $E_{\text{VBM,Ref}}^B$ and $E_{\text{VBM,Ref}}^A$ are the energies of the valence band maximum (VBM) of bulk materials B and A, respectively, measured relative to a common reference level, such as the average electrostatic potential. $\Delta\bar{V}^{AB}$ is the average electrostatic potential difference between the two sides of the interface.

The CBO was calculated using the same approach as

$$\text{CBO}(A/B) = E_{\text{CBM,Ref}}^B - E_{\text{CBM,Ref}}^A + \Delta\bar{V}^{AB}, \quad (2)$$

where $E_{\text{CBM,Ref}}^B$ and $E_{\text{CBM,Ref}}^A$ are the energies of the conduction band minimum (CBM) of bulk materials B and A, respectively.

To investigate the dependence of the bandgap on the alloy composition, $1 \times 3 \times 2$ special quasi-random structure (SQS) supercells were employed. Two- and three-atom clusters are used to minimize the correlation mismatch between SQS and truly random alloy structures, as implemented in the *mcSQS* code distributed with the Alloy Theoretic Automated Toolkit [31]. Al cations preferentially occupy octahedral sites [6,7]. Accordingly, for the generation of SQS at Al concentrations below 50% (i.e., $x < 0.5$), Al cations are randomly substituted among the octahedral sites. For Al concentration exceeding 50%, all octahedral sites become fully occupied, and the remaining Al cations are randomly substituted into the tetrahedral sites. For $(\text{Al}_{0.5}\text{Ga}_{0.5})_2\text{O}_3$ film, an additional superlattice structure was employed in which all octahedral sites of the film are occupied by Al cations. The values of the average electrostatic potential differences, $\Delta\bar{V}^{AB}$, for $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ alloys are obtained by means of linear interpolation.

β -($\text{Al}_x\text{Ga}_{1-x}$) $_2\text{O}_3$ / Ga_2O_3 device simulations were performed using Applied Materials' Ginestra® [32], a state-of-the-art simulation software that provides a comprehensive description of the transport in electronic devices. In Ginestra, all the relevant charge transport mechanisms are simultaneously accounted for (including drift-diffusion in the materials' bands, thermionic emission, direct tunneling, Fowler-Nordheim tunneling, and multiphonon trap-assisted tunneling), with the relative equations self-consistently solved together with Poisson's equation. The simulation activity focused on generating the ideal current-voltage (I - V) and C - V characteristics of Pt/β -($\text{Al}_x\text{Ga}_{1-x}$) $_2\text{O}_3$ / Ga_2O_3 SBD, considering the parameters provided by the *ab initio* DFT calculations presented in this study. Comparisons with previously published experimental I - V curves are also presented, validating the presented DFT calculation results. The main parameters

used for the simulations are listed in Table S1 in Supplemental Material [33–36].

III. RESULTS AND DISCUSSION

A. Pseudomorphic Al₂O₃ films on β -Ga₂O₃ substrates

Because the lattice mismatch between Al₂O₃ films and the Ga₂O₃ substrates, an in-plane tensile strain is induced during the Al₂O₃ growth. For each pseudomorphic growth direction—namely, (100)B, (010), (001)B, and $\bar{2}$ (01)—the in-plane lattice parameters of the Al₂O₃ films were matched to those of the β -Ga₂O₃ substrate, while the structures were relaxed along the out-of-plane direction. We refer to these strained Al₂O₃ structures as the “films.” The in-plane tensile strain of the films causes significant shifts in the Al₂O₃ band structure. The magnitude of these shifts can be evaluated from the bandgaps, VBM, and CBM as reported in Table I. In the same table, the lattice parameters for bulk β -Ga₂O₃, θ -Al₂O₃, and Al₂O₃ films are also reported.

Note that the CBM and VBM energy reported in Table I are those used in Eqs. (1) and (2) as reference energy. That is because the film structures have been relaxed independently, i.e., before forming an interface with the β -Ga₂O₃ substrate. On the other hand, the average electrostatic potential difference, $\Delta\bar{V}^{AB}$, has been computed after the interface has been formed, i.e., after full relaxation of the heterostructure. It is evident from the energies reported in Table I that the strain yields a notable effect on the bandgaps of the Al₂O₃ films, which is smaller than that of bulk θ -Al₂O₃. This is also true for the CBM and VBM of the films, although the variation of the VBM is less pronounced. Given these sizeable variations of the reference energies, it is crucial to relax the film structure before

computing the CBO and VBO using Eqs. (1) and (2). Among the studied orientations, (100)B exhibits the smallest band-edge shifts, originating from its lower net strain (and thus smaller volume change, see Table I) compared with other orientations. As a consequence, the change in bandgap is also minimal.

The role of the strain in determining the band offset in heterostructures is also shown in Ref. [13]. There, two different substrate–film combinations were used: Ga₂O₃ as the substrate with an Al₂O₃ film or Al₂O₃ as the substrate with a Ga₂O₃ film. In both cases, the films’ in-plane lattice parameters were constrained to match those of the respective substrate lattice. Results of Ref. [13] show that swapping substrates with films not only affects the magnitude of the band offset but also changes the type of band offset, as the VBO changes sign from -0.14 eV (when β -Ga₂O₃ is the substrate) to 0.54 eV (when Al₂O₃ is the substrate).

Figure S2 in Supplemental Material [33] illustrates the θ -Al₂O₃/ β -Ga₂O₃ superlattice structures for the (100)B, (010), (001)B, and $\bar{2}$ (01) orientations. As previously discussed, only nonpolar, symmetric, and most stable terminations were employed to prevent any built-in potential difference at the interfaces. The in-plane lattice parameters were fixed to those of the substrate, while the out-of-plane lattice parameters of the superlattice structures were relaxed to release the strain along that direction. After the cell relaxations, full atomic relaxation at a fixed cell structure was performed.

The averaged electrostatic potential profiles are presented in Fig. S4 in Supplemental Material [33]. The planar average electrostatic potential is computed by averaging the electrostatic potential over the in-plane directions, while the macroscopic average is computed along the out-of-plane direction. An essentially constant macroscopic average indicates bulk-like behavior, confirming the absence of any built-in potential at the interfaces. A constant macroscopic average potential serves as a reference for the band offset calculations. The alignment procedure described in Eqs. (1) and (2) is then applied to obtain the band offset. Figure 1 illustrates the VBO and CBO of the Ga₂O₃/Al₂O₃ heterojunctions, highlighting their dependence on different interface orientations, namely, (100)B, (010), (001)B, and $\bar{2}$ (01).

The calculated values vary significantly across orientations, not only in magnitude but also in type. For instance, the $\bar{2}$ (01) orientation exhibits a band offset (type I straddling) different from that of the other cases. The (100)B orientation shows the largest VBO and CBO, equal to 0.53 and 2.44 eV, respectively. In contrast, the $\bar{2}$ (01) orientation displays the smallest offsets, with a VBO of only -0.05 eV. The VBO reported for $\bar{2}$ (01) aligns closely with those experimentally determined in Ref. [19] by means of XPS for films prepared by ALD. The discrepancy between the CBO shown in Fig. 1 and the one

TABLE I. Calculated lattice parameters (a, b, c) and volume per formula unit ($V_{f.u.}$) obtained using the SCAN functional. The bandgaps (E_g) and relative positions of the $E_{CBM,Ref}$ and $E_{VBM,Ref}$ are determined using the HSE06 hybrid functional for Ga₂O₃, unstrained bulk θ -Al₂O₃ and strained θ -Al₂O₃ bulk corresponding to the most common crystal growth orientations. The strained in-plane lattice constants are denoted as a and b , while the relaxed out-of-plane constant is c . The transformation matrix for $\bar{2}$ (01) orientation is provided in Supplemental Material [33].

	β -Ga ₂ O ₃	θ -Al ₂ O ₃	(100)B	(010)	(001)B	$\bar{2}$ (01)
a	12.21	11.77	3.04	5.83	12.21	3.04
b	3.04	2.90	5.83	12.21	3.04	14.71
c	5.83	5.61	11.31	2.87	5.52	5.53
$V_{f.u.}$	52.54	46.48	48.64	49.47	49.75	49.95
E_g	4.71	7.19	6.62	6.49	6.51	6.45
$E_{CBM,Ref}$	6.875	11.022	9.997	9.634	9.651	9.580
$E_{VBM,Ref}$	2.17	3.832	3.378	3.140	3.146	3.133

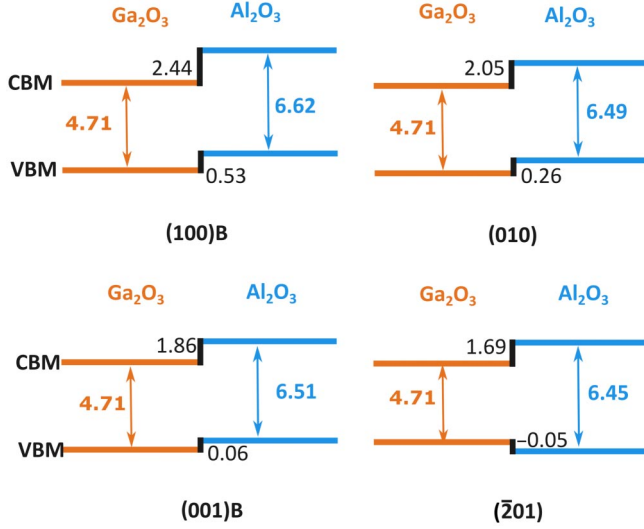


FIG. 1. Calculated band offsets (in eV) between $\beta\text{-Ga}_2\text{O}_3$ and $\theta\text{-Al}_2\text{O}_3$ for (100)B, (010), (001)B, and $(\bar{2}01)$ orientations.

reported in Ref. [19] is due to the experimental value of the bandgap (6.9 eV), as only the VBO is measured directly. These results agree with the trend observed in previous modeling work [18]. CBO for (010) and (001) orientations is almost identical to the values (2.24 and 1.86 eV, respectively) reported for strained slabs [18]. This very close agreement is likely due to almost ignorable, in-plane-only, atomic relaxation observed in slab geometries for the (010) orientation [7]. Notable differences between our results and those reported in Ref. [18] are found for other orientations. These differences suggest that band offset calculations done using separate slab geometries are not always adequate, as they fail to model the chemical bonding at the interface between films and substrates. This failure is particularly evident when a non-negligible out-of-plane atomic displacement at the interface occurs [7], which can substantially lower the electronic potential at the interface. We note that domain mixing among up to three competing strained variants, as recently predicted for heterostructures with monoclinic crystal systems [37], can provide an additional source of band offset broadening, at least for the $(h0l)$ orientations.

B. Pseudomorphic $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ films on $\beta\text{-Ga}_2\text{O}_3$ substrates

The bandgaps of strained $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ films pseudomorphically grown on $\beta\text{-Ga}_2\text{O}_3$ substrates have been calculated as a function of the Al content for each of the above-mentioned orientations. An increase in the Al content always yields a widening of the bandgap, as expected, since the bandgap of Al_2O_3 is larger than that of Ga_2O_3 . However, a non-negligible deviation from linearity is observed from a positive value of the bowing parameter, b , determined by fitting the relation:

$$E_g(x) = (1-x)E_g[\text{Ga}_2\text{O}_3] + xE_g[\text{Al}_2\text{O}_3] - bx(1-x), \quad (3)$$

where $E_g[\text{Ga}_2\text{O}_3]$ and $E_g[\text{Al}_2\text{O}_3]$ are the bandgaps of the pure compounds, $\beta\text{-Ga}_2\text{O}_3$ and strained $\theta\text{-Al}_2\text{O}_3$. The bowing parameters for $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ alloys are generally around 1 eV, in agreement with previously reported values [14]. Only in the case of the (100) direction, a slightly higher value of 1.2 eV is observed.

The knowledge of the absolute positions of the VBM and CBM of the alloys is useful in view of applications, as these values are measured as ionization energy and electron affinity. To assess the absolute VBM and CBM, we conducted additional supercell calculations along the out-of-plane direction, with a vacuum. We refer to this calculation as surface structures. The results are shown in Fig. 2 for $\beta\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$ alloys.

Each subplot shows the variation of CBM and VBM energies as a function of Al content, x , illustrating how band-edge positions shift with increasing Al concentration. Absolute CBM and VBM of $\beta\text{-Ga}_2\text{O}_3$ are also shown for comparison. Notably, there is a significant orientation-dependent variation in the absolute CBM and VBM energies. For instance, the CBM for (001)B and $(\bar{2}01)$ orientations lies at energies higher than for (010) and (100)B orientations, suggesting a strong influence of the film growth direction on the electron affinity.

The observed trend suggests that Al substitution primarily affects the conduction band offset, increasing the electron affinity of the alloy without altering the position of the valence band significantly. This trend is likely due to the dominance of O 2p states at the VBM, which are less sensitive to the change in cation composition between Ga and Al. This increase in CBO with Al content could enhance electron confinement in heterojunction structures, which is beneficial for device applications. Such orientation-dependent behavior could be leveraged in device applications to tailor the band offset properties at heterojunction interfaces, depending on the desired electronic characteristics.

C. Schottky barrier diode modeling

To investigate the practical effects of the $\text{Ga}_2\text{O}_3/(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ interface orientation on a device operation, we used the Ginestra® simulation software to reproduce the I - V and C - V characteristics of Pt/ $\beta\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3/\text{Ga}_2\text{O}_3$ SBD structures with 25% Al content, using the calculated materials parameters for the four orientations $(\bar{2}01)$, (010), (100), and (001).

As clearly visualized in the band diagrams of the simulated SBD [Fig. 3(b)], the interface orientation affects both the $\text{Ga}_2\text{O}_3/(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ conduction band offset—as expected due to the independent variation of the two materials' electron affinities—and the $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3/\text{Pt}$ Schottky barrier height (SBH)—the Pt work function

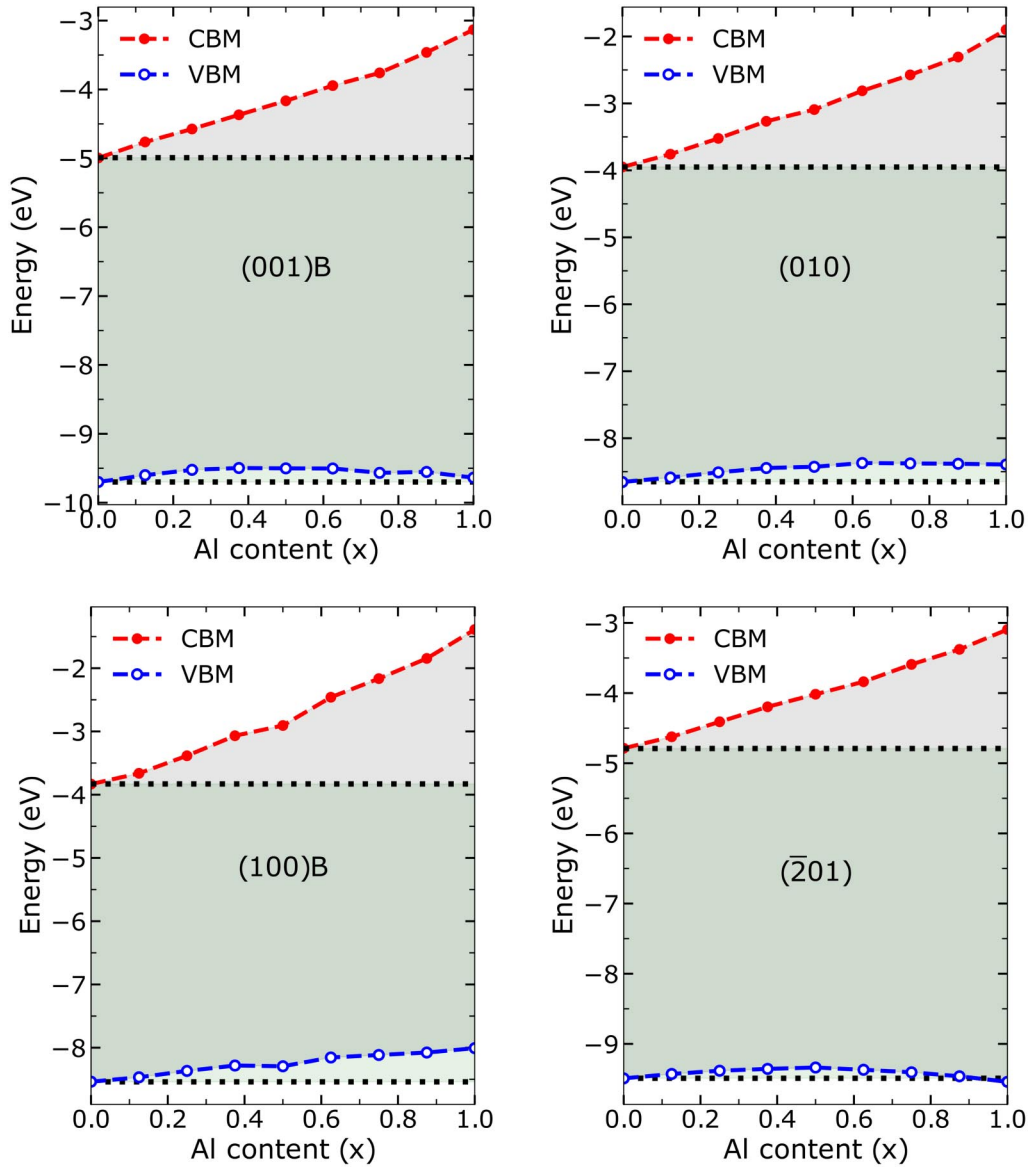


FIG. 2. Computed absolute energies of the CBM and VBM (in eV) using superlattice structure calculations for β -(Al_xGa_{1-x})₂O₃. The horizontal black dotted lines represent the CBM and VBM of β -Ga₂O₃. The reported values are relative to the vacuum level, determined using a vacuum slab.

being unaffected by the orientation. Figure 3(a) shows the I - V characteristics of the simulated SBD for the four considered orientations.

At negative bias (i.e., the reverse bias condition for the SBD), the charge transport is given by a combination of thermionic emission and tunneling processes [38] from the Pt cathode at the Pt/ β -(Al_xGa_{1-x})₂O₃ interface, with both the mechanisms strongly and inversely dependent on the Pt/ β -(Al_{0.25}Ga_{0.75})₂O₃ SBH. This is consistent with the simulation results shown in Fig. 3, where a lower current magnitude corresponds to a higher Pt/ β -(Al_{0.25}Ga_{0.75})₂O₃ SBH and thus to a lower electron affinity of the specific (Al_{0.25}Ga_{0.75})₂O₃ orientation. Further Ginestra® simulations allowed us to inspect the specific nature of the

negative bias charge transport by running different simulations while enabling a single transport mechanism at a time. For all four simulated (Al_{0.25}Ga_{0.75})₂O₃/Ga₂O₃ interface orientations, the negative bias transport results were dominated by the tunneling through the evolving trapezoidal/triangular (Al_{0.25}Ga_{0.75})₂O₃ barrier.

At positive bias (i.e., the forward bias condition for the SBD), the charge transport was also investigated by using Ginestra® and applying the same methodology used for the negative bias region. This results in a combination of diffusion current and thermionic emission at low bias, followed by a combination of thermionic emission and tunneling processes at higher positive bias. The contribution related to the diffusion process is recognizable in the

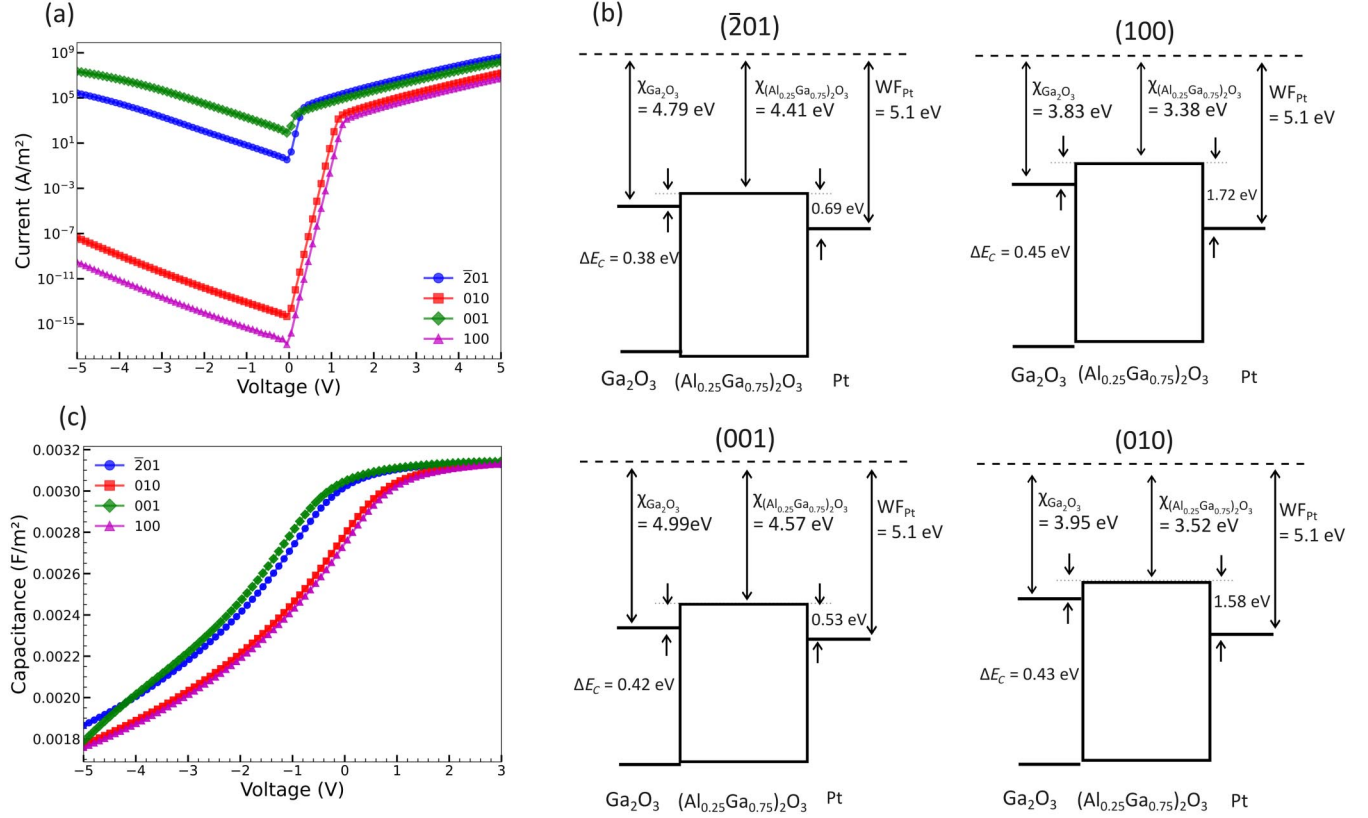


FIG. 3. (a) Simulated I - V characteristics of $\text{Pt}/(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3/\text{Ga}_2\text{O}_3$ SBDs for different orientations: $\bar{2}01$, 010 , 001 , and 100 . The absolute value of the current is shown in the logarithmic vertical scale. (b) Energy band diagrams for the four orientations, illustrating work function (WF_{Pt}), electron affinity (χ), and conduction band offset (ΔE_C). (c) Simulated C - V characteristics for the same orientations, highlighting variations in capacitance with applied voltage. The $(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3$ relative dielectric permittivity is set to 11.15, and the thickness of the layer is set to 30 nm for all simulations.

low-voltage region, most noticeably for the 100 and 010 orientations, characterized by the exponential increase in the current with the ideal slope of $q/\ln(k_B T) = 60$ mV/decade at room temperature. This originates from the presence of a depletion region in the Ga_2O_3 due to the built-in voltage V_{bi} of the SBD (i.e., the difference between the Ga_2O_3 electron affinity and the Pt work function) imposed by the specific orientation. Noticeably, the presence of a depletion region barrier in the Ga_2O_3 is the main bottleneck for the majority carriers' transport at low field. Upon the application of a positive bias, the barrier is progressively lowered, leading to an exponential increase in the current due to majority carriers' diffusion over the Ga_2O_3 barrier.

As the Ga_2O_3 reaches flatband ($V = V_{\text{bi}}$) or accumulation condition ($V > V_{\text{bi}}$), with no depletion region barrier in place, the charge transport becomes once again limited by the thermionic emission and tunneling through an evolving trapezoidal/triangular $(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3$ barrier.

The voltage dependency of the limiting transport mechanism is clearly visualized in the I - V characteristics of Fig. 3(a): at low voltage, the current is characterized by

an exponential increase, transitioning into a more complex trend at high voltage. In the low-voltage regime, the current magnitude is governed by the Ga_2O_3 electron affinity, while at high voltage, it is determined by the $(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3/\text{Ga}_2\text{O}_3$ conduction band offset.

From the presented analysis, we note that the specific $(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3/\text{Ga}_2\text{O}_3$ orientation can be used for tailoring the performance of the devices. For example, using the 100 orientation results in an SBD with an extremely high $I_{\text{ON}}/I_{\text{OFF}}$ ratio, while the $\bar{2}01$ orientation results in an SBD characterized by a low built-in voltage (thus fast turn-on) and higher forward current.

The simulated high-frequency (1 MHz) C - V characteristics of the same SBD are shown in Fig. 3(c), exhibiting very similar behaviors regardless of the specific orientation. The simulated C - V characteristics resemble those of a typical MOS capacitor, with a deep-depletion trend at negative voltage and with the maximum capacitance at positive bias voltage (i.e., in nominal accumulation) determined by the $(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3$ layer dielectric constant and thickness. The only significant difference between the simulated C - V characteristics is a voltage shift given by the orientation-dependent built-in voltage.

The simulated C - V characteristics are remarkably similar to those previously reported for (Al_{0.08}Ga_{0.92})₂O₃/Ga₂O₃ modulation-doped field-effect transistors [39], presenting a fundamental MOS structure very similar to the ones presented and simulated in this work. It is worth noting that the reported (Al_{0.08}Ga_{0.92})₂O₃/Ga₂O₃ structure is capable of confining a two-dimensional electron gas at the interface, and, given the similarities with our simulated structures, we can speculate that they can also exhibit such a capability.

The experimental I - V measurements provided in Refs. [34,36] were used to validate our simulations. In particular, we want to assess how accurately device simulations based on band offsets and dielectric constants from first principles can match published experimental I - V data. Values of the band offset and electron affinity (3.60 eV) for 21% Al content are obtained by interpolation of the first-principles results shown in Table S2 in Supplemental Material [33]. These values are used in the device simulations to generate I - V characteristics for comparison against the experimental results reported by Mudiyansele *et al.* [36]. The simulation is performed for an ideal Pt/(Al_{0.21}Ga_{0.79})₂O₃/Ga₂O₃ structure in the absence of any defect profiles. The thickness of the (Al_{0.21}Ga_{0.79})₂O₃ layer is set to 200 nm for the interface oriented in the (010) direction. Additional simulation parameters are listed in Table S1 in Supplemental Material [33]. The simulated and experimental I - V curves are plotted in Fig. 4.

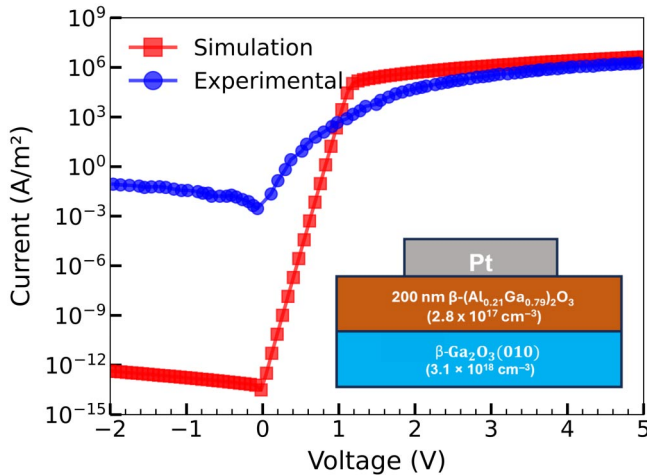


FIG. 4. Comparison of simulated (ideal) and experimental I - V data from Ref. [36] for (Al_{0.21}Ga_{0.79})₂O₃/Ga₂O₃ (010) SBD. The thickness of the (Al_{0.21}Ga_{0.79})₂O₃ layer (200 nm) in the simulation is identical to the (Al_{0.21}Ga_{0.79})₂O₃ thickness used in the device of Ref. [36]. The electron affinity values [3.60 eV for (Al_{0.21}Ga_{0.79})₂O₃ and 3.95 eV for Ga₂O₃ (010)] were interpolated from Table S2 in Supplemental Material [33]. The inset shows a schematic of the simulated device with the doping concentrations for each layer.

The trend is well reproduced, with discrepancies in the current magnitude. At low positive bias, the experimental curve deviates from the simulated characteristic, likely due to the presence of defects at the (Al_{0.21}Ga_{0.79})₂O₃/Ga₂O₃ interface, which are not accounted for in our simulations. Interface defects are known to induce higher current through the Shockley-Read-Hall carrier generation and recombination mechanisms.

The experimental forward current does not present a sharp transition from the diffusion to the tunneling regime, possibly indicating a heterojunction potential less abrupt than the simulated one or a surface potential pinning effect due to the presence of interface states at the Ga₂O₃/(Al_{0.21}Ga_{0.79})₂O₃ interface. The discrepancy between the experimental and simulated I - V in the negative bias region is likely caused by defects in the bulk of the (Al_{0.21}Ga_{0.79})₂O₃ layer.

The first principles calculated band offset parameters were used to simulate SBD I - V characteristics and compare them to previously published experimental I - V curves (Figs. 4 and 6) [34,36]. The simulations matched remarkably well with the experimental data in a bias regime where the current is most sensitive to the band offset values, thereby validating the first-principles calculations. Matching the experimental I - V s across the full bias range is outside the scope of this work. However, by considering a defect band in the (Al_xGa_{1-x})₂O₃ layer (donor defect 1.38 eV below the (Al_xGa_{1-x})₂O₃ conduction band) aligned with the Pt Fermi level, it is possible to clearly illustrate how the SBD current at reverse bias is affected by the presence of defects (Fig. 5).

Figure 6 shows the experimental I - V characteristics reported by Sundaram *et al.* [34], along with the simulated I - V curves generated in this study. The I - V curves are obtained for an SBD fabricated on MOCVD grown (Al_{0.22}Ga_{0.78})₂O₃ on β -Ga₂O₃ (010) substrate. The doping concentration in the β -Ga₂O₃ substrate and in the 30 nm (Al_{0.22}Ga_{0.78})₂O₃ layer are assumed to be $3 \times 10^{16} \text{ cm}^{-3}$, identical to the doping level reported in Ref. [34]. The differences in the I - V characteristics between Figs. 4 and 6 arise from the varying (Al_xGa_{1-x})₂O₃ layer thicknesses and doping concentrations (see Fig. S5 in Supplemental Material [33]).

In the forward bias region, Fig. 6 shows a very good agreement between the simulation and the published experimental data. As for the simulation reported above, the simulated I - V curve is obtained for an ideal case, in the absence of any defect profile in the structure. It is interesting to note that in the forward region, the experimental I - V curve shows a voltage dependence that follows the ideal curve more closely than in the previous case, as reported in Fig. 4. We also note that the I - V profile presents a sharper transition from diffusion to thermionic regimes, suggesting a more abrupt (Al_{0.22}Ga_{0.78})₂O₃/Ga₂O₃ interface for the device of Ref. [34] than that of

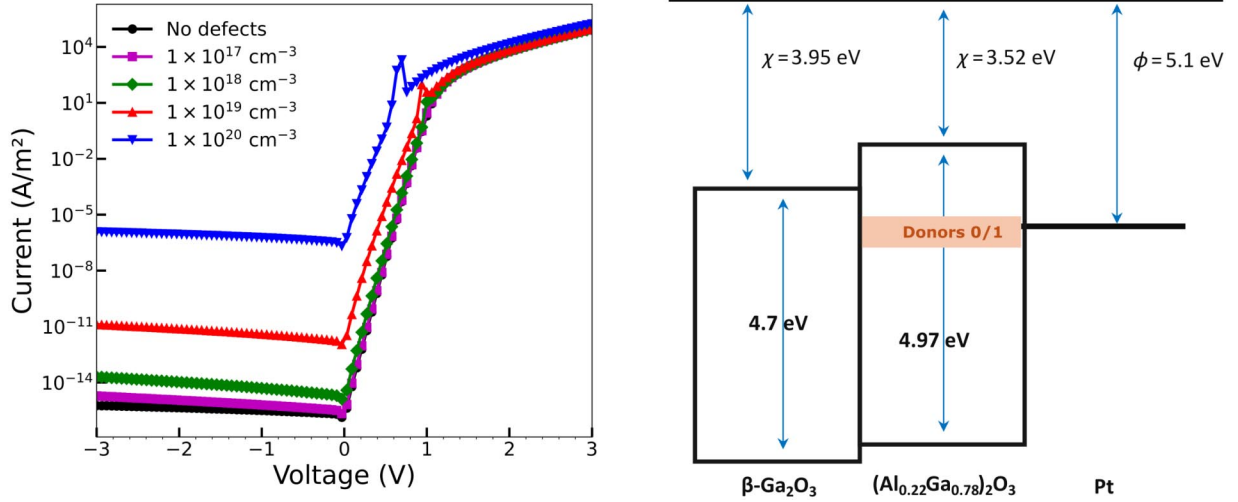


FIG. 5. Simulated I - V curves for $(\text{Al}_{0.22}\text{Ga}_{0.78})_2\text{O}_3/\text{Ga}_2\text{O}_3$ (010) SBD considering different defect concentrations in the $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ layer. Direct and trap-assisted tunneling conduction mechanisms are included in the simulation.

Ref. [36]. The main differences between the experimental I - V curve and the simulated data observed in Fig. 6 are the horizontal shift, possibly due to a mismatch in the built-in voltage or the presence of negative charge in the materials' stack. As previously discussed, the positive shift in the experimental I - V could be caused by either (i) an experimental Schottky barrier larger than the ideal value (i.e., the difference between the Pt work function and the Ga_2O_3 Fermi level) considered in the simulations or (ii) the presence of negative charge and depletion at the $(\text{Al}_{0.22}\text{Ga}_{0.78})_2\text{O}_3/\text{Ga}_2\text{O}_3$ interface. The higher reverse

current in the negative bias region probably has a similar origin to the previous example, caused by defects that increase the Schottky reverse leakage current.

IV. CONCLUSION

We have conducted computational modeling using the Heyd-Scuseria-Ernzerhof hybrid functional and technology computer-aided design models to investigate band offset, I - V , and C - V for different growth orientations and alloy compositions in $\text{Ga}_2\text{O}_3/(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ heterojunctions. Strain from lattice mismatch affects both VBM and CBM, with distinct behaviors observed across $(\bar{2}01)$, (010), (001), and (100) orientations due to the high anisotropy of the monoclinic phase. We examined the effect of the strain on band offset by incorporating its effect on bulk energy levels and the potential lineup at the interface to ensure a consistent reference. The high sensitivity to growth orientation and strain may justify the discrepancies in previously published results. Additionally, we report C - V and I - V simulations for $\text{Ga}_2\text{O}_3/(\text{Al}_{0.25}\text{Ga}_{0.75})_2\text{O}_3/\text{Pt}$ SBD across the four growth orientations, revealing significant orientation dependence given the different materials' electron affinities and the resulting conduction band offsets. To validate our findings, we simulated and compared with experimental I - V results for Al concentrations of 22% and 21%, achieving reasonable agreement in the forward bias. These results provide insights into the device structure and the potential defect and highlight the critical role of orientation and strain engineering in optimizing wide-bandgap devices for power electronics.

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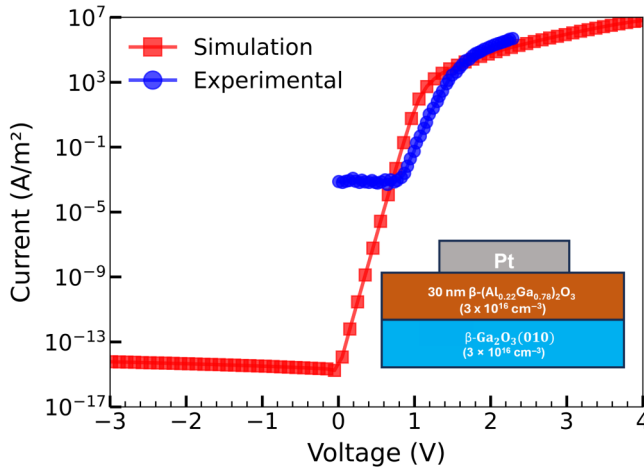


FIG. 6. Comparison of simulated (ideal) and experimental I - V data from Ref. [34] for $(\text{Al}_{0.22}\text{Ga}_{0.78})_2\text{O}_3/\text{Ga}_2\text{O}_3$ (010) SBD. The interpolated electron affinity value of $(\text{Al}_{0.22}\text{Ga}_{0.78})_2\text{O}_3$ (010) is 3.58 eV and Ga_2O_3 (010) is 3.95 eV (Table S2 in Supplemental Material [33]). The $(\text{Al}_{0.22}\text{Ga}_{0.78})_2\text{O}_3$ thickness and mobility values considered for the simulation are 30 nm and $184 \text{ cm}^2 \times \text{V}^{-1} \times \text{s}^{-1}$, respectively (identical to the values provided by Ref. [34]). The inset shows a schematic of the simulated device with the doping concentrations for each layer.

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DATA AVAILABILITY

Computational parameters are provided in the main manuscript and Supplemental Material [33]. Additional data are available from the corresponding author upon reasonable request.

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