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# Growth of conductive Si-doped $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> by suboxide molecular-beam epitaxy

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## ABSTRACT

We report a two-step film-growth process using suboxide molecular-beam epitaxy (S-MBE) that produces Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> with record transport properties. The method involves growing a relaxed  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layer on *m*-plane sapphire at a relatively high substrate temperature ( $T_{\text{sub}}$ ),  $\sim 750$  °C, followed by an Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> overlayer grown at lower  $T_{\text{sub}}$ ,  $\sim 500$  °C. The high  $T_{\text{sub}}$  allows the  $\sim 3.6\%$  lattice-mismatched  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer with  $x = 0.08 \pm 0.02$  to remain epitaxial and phase pure during relaxation to form a pseudosubstrate for the overgrowth of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>. The optimal conditions for the subsequent growth of Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> by S-MBE are  $425$  °C  $\leq T_{\text{sub}} \leq 500$  °C and  $P_{80\% \text{ O}_3} = 5 \times 10^{-6}$  Torr. Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown with this method at  $T_{\text{sub}} > 550$  °C are always insulating. Secondary-ion mass spectrometry confirms that both the insulating and conductive films have uniform silicon incorporation. In conductive films with  $10^{19} \leq N_{\text{Si}} \leq 10^{20}$  cm<sup>-3</sup>, the incorporated silicon is  $\sim 100\%$  electrically active. At  $N_{\text{Si}} \leq 10^{19}$  cm<sup>-3</sup>, the carrier concentration ( $n$ ) plummets. A maximum Hall mobility ( $\mu$ ) =  $90 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  at room-temperature is measured in a film with  $n = 2.9 \times 10^{19}$  cm<sup>-3</sup> and a maximum conductivity ( $\sigma$ ) = 650 S/cm at room-temperature in a film with  $n = 4.8 \times 10^{19}$  cm<sup>-3</sup>. A threading dislocation density of  $(5.6 \pm 0.6) \times 10^{10}$  cm<sup>-2</sup> is revealed by scanning transmission electron microscopy, showing that there is still enormous room to improve the electrical properties of doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films.

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## INTRODUCTION

One of the most important metrics for wide and ultrawide bandgap semiconductors is the Baliga figure of merit (BFOM), which describes the theoretical minimum on-resistance for a given breakdown field of an ideal power rectifier made of a specific material.<sup>1</sup> The BFOM is derived to be equal to  $E_c^3 \mu \epsilon_s$ , where  $E_c$  is

the maximum electric field that can be applied to a semiconductor before breakdown,  $\mu$  is the mobility, and  $\epsilon_s$  is the permittivity of the semiconductor.<sup>1</sup> Wider bandgaps greatly increase the BFOM because  $E_c$  scales with  $E_g^{-1.83}$ , where  $E_g$  is the bandgap of the semiconductor, resulting in the BFOM scaling with  $E_g^{-5.5}$  and being dominated by ultrawide bandgap materials, such as Ga<sub>2</sub>O<sub>3</sub>, AlN, and diamond.<sup>2-4</sup> Importantly, to be considered an ultrawide bandgap

semiconductor rather than an insulator, it is necessary to be able to introduce mobile carriers via doping to modulate the conductivity. Of the primary UWBG semiconductors, Ga<sub>2</sub>O<sub>3</sub> has the narrowest bandgap of 4.6–5.4 eV, depending on the Ga<sub>2</sub>O<sub>3</sub> polymorph. Until now, β-Ga<sub>2</sub>O<sub>3</sub> has received significant attention since it has controllable *n*-type doping, room temperature electron mobilities as high as 200  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$ , and a projected critical field of 8 MV/cm and is the only ultrawide bandgap semiconductor with large-diameter (>4 in) native substrates.<sup>4,5</sup>

In addition to the thermodynamically stable monoclinic β phase, there are four additional phases in the Ga<sub>2</sub>O<sub>3</sub> system: trigonal (α), cubic (γ and δ), and orthorhombic (κ).<sup>1–3</sup> Of these, the metastable α-Ga<sub>2</sub>O<sub>3</sub> phase has the widest bandgap of 5.4 eV.<sup>6</sup> The ultrawide bandgap of α-Ga<sub>2</sub>O<sub>3</sub> can be further extended to 8.6 eV by alloying with α-Al<sub>2</sub>O<sub>3</sub>, also increasing the breakdown field to an anticipated 30 MV/cm.<sup>2,7</sup> Both the bandgap and breakdown field exceed those of all known semiconductors. In addition, α-Ga<sub>2</sub>O<sub>3</sub> has an electron effective mass of only  $m_e^* = 0.276 m_e^0$ , where  $m_e^0$  is the mass of a free electron and  $m_e^*$  is the effective mass of an electron in α-Ga<sub>2</sub>O<sub>3</sub>. This is significantly lower than the  $m_e^* = 0.342 m_e^0$  of β-Ga<sub>2</sub>O<sub>3</sub>.<sup>8</sup> The other compositional extreme, α-Al<sub>2</sub>O<sub>3</sub>, has an even lower reported electron effective mass of  $m_e^* = 0.23 m_e^0$ , leading to a relatively high predicted electron mobility.<sup>9</sup>

It is predicted that the room-temperature mobility of α-Ga<sub>2</sub>O<sub>3</sub> is limited by polar-optical phonons for  $n \geq 10^{15} \text{ cm}^{-3}$ , just as it is for β-Ga<sub>2</sub>O<sub>3</sub>.<sup>6</sup> The maximum mobility value at room temperature for α-Ga<sub>2</sub>O<sub>3</sub> is predicted to be  $\sim 220 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$ , around 10% higher than the 200  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  maximum predicted and observed for β-Ga<sub>2</sub>O<sub>3</sub>.<sup>6,10,11</sup> Achieving the maximum mobility for α-Ga<sub>2</sub>O<sub>3</sub> is expected to primarily be a challenge of finding optimal growth (including an appropriate substrate or pseudosubstrate) and doping conditions to maximize crystallinity.<sup>6,12</sup> For α-Al<sub>2</sub>O<sub>3</sub>, the theoretical room-temperature mobility is as high as 600  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  and the theoretical low-temperature mobility is as high as 30 000  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$ .<sup>8,13</sup> Given the record bandgaps and breakdown fields, as well as the high predicted mobilities, the α-(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> system has the potential to achieve the highest BFOM of any known semiconducting material.

Many methods have been utilized for the growth of α-(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> epitaxial films, including hydride vapor-phase epitaxy (HVPE),<sup>14–16</sup> mist chemical vapor deposition (mist CVD),<sup>17–20</sup> molecular-beam epitaxy (MBE),<sup>7,21,22</sup> metalorganic chemical vapor deposition (MOCVD),<sup>23,24</sup> and pulsed-laser deposition (PLD).<sup>25,26</sup> Using suboxide MBE (S-MBE) on *a*-plane sapphire, we previously demonstrated the growth of α-(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> with  $0 < x < 0.95$  at growth rates as high as 2.9 μm/h and with the narrowest average symmetric rocking curve full width at half maximum (FWHM) of any study, 12 arc sec.<sup>22</sup> On *c*-plane sapphire, the highest quality α-Ga<sub>2</sub>O<sub>3</sub> has been demonstrated by mist CVD and shows symmetric rocking curves with FWHM as fine as 25 arc sec.<sup>17</sup> On *m*-plane sapphire, HVPE has demonstrated the highest quality α-Ga<sub>2</sub>O<sub>3</sub>, with symmetric rocking curve FWHM as low as 1000 arc sec at growth rates of up to 6 μm/h.<sup>16</sup> MBE has not demonstrated phase-pure, thick α-Ga<sub>2</sub>O<sub>3</sub> films on *c*-plane sapphire and has demonstrated a relatively large symmetric rocking curve FWHM of 1600 arc sec on *m*-plane sapphire.<sup>7,27</sup> For MBE, the *a*-plane and *c*-plane both have limited critical thicknesses for phase-pure α-Ga<sub>2</sub>O<sub>3</sub> before a transition to β-Ga<sub>2</sub>O<sub>3</sub> occurs.<sup>7,28</sup> On *m*-plane sapphire, conventional

MBE has demonstrated growth across the full range of *x* with no critical thickness for phase-pure α-Ga<sub>2</sub>O<sub>3</sub>.<sup>7,29</sup> More recently, applying metal-oxide catalyzed epitaxy (MOCATAXY) to MBE growth of α-Ga<sub>2</sub>O<sub>3</sub> on *m*-plane sapphire extended the growth rate up from 0.12 to 0.2 μm/h while also reducing the FWHM of the 300 α-Ga<sub>2</sub>O<sub>3</sub> rocking curves from 1800 to 1600 arc sec.<sup>27</sup> For reasons that are not yet understood, it has also been seen in mist CVD that the mobilities of Sn-doped α-Ga<sub>2</sub>O<sub>3</sub> grown on *m*-plane sapphire are significantly higher than films grown in similar conditions on *c*-plane sapphire despite worse crystallinity and similar levels of impurities.<sup>30</sup>

For the doping of α-Ga<sub>2</sub>O<sub>3</sub>, there are a wide variety of shallow *n*-type donors predicted by density functional theory (DFT), including silicon, hafnium, zirconium, tin, tantalum, and germanium.<sup>31</sup> With tin and silicon alone, many methods including mist CVD, PLD, MBE, MOCVD, and HVPE have successfully demonstrated conductivity in doped α-Ga<sub>2</sub>O<sub>3</sub>.<sup>15,17,24,26,28,30</sup> The highest reported α-Ga<sub>2</sub>O<sub>3</sub> conductivity is 37 S/cm in an Sn-doped film with a mobile electron concentration of  $1.4 \times 10^{19} \text{ cm}^{-3}$  grown using PLD, and the record for mobility is 65  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  in an Sn-doped film with a mobile electron concentration of  $1.2 \times 10^{18} \text{ cm}^{-3}$  grown by mist CVD.<sup>23,27</sup> Doping with silicon has achieved a maximum mobility of 51.6  $\frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  at a mobile electron concentration of  $1.7 \times 10^{18} \text{ cm}^{-3}$  in α-Ga<sub>2</sub>O<sub>3</sub> grown using HVPE.<sup>15</sup> We view silicon as the most promising dopant since it is expected to remain a shallow donor up to 72% aluminum incorporation, the highest of any known dopant.<sup>31</sup>

The limited mobilities and conductivities in doped α-Ga<sub>2</sub>O<sub>3</sub> thin films are believed to be primarily due to traps from high dislocation and impurity densities. The typical substrate for growing α-Ga<sub>2</sub>O<sub>3</sub> is corundum α-Al<sub>2</sub>O<sub>3</sub> as it is high quality and isostructural, but α-Al<sub>2</sub>O<sub>3</sub> has a significant compressive lattice mismatch to α-Ga<sub>2</sub>O<sub>3</sub> of −3.54% along the *c*-axis and −4.81% along the *a*-axis.<sup>32</sup> In >100 nm thick films, this mismatch results in the generation of  $10^{10}$ – $10^{11} \text{ cm}^{-2}$  threading dislocations during the structural relaxation of α-Ga<sub>2</sub>O<sub>3</sub> from the α-Al<sub>2</sub>O<sub>3</sub> substrate.<sup>27,32–34</sup> Such high dislocation densities cause the mobility to become dislocation-limited at carrier concentrations of  $10^{19} \text{ cm}^{-3}$  and lower.<sup>35</sup> For doping at concentrations of  $\leq 10^{16} \text{ cm}^{-3}$ , as are relevant for high-voltage vertical diodes, the dislocation density must be reduced to  $10^8 \text{ cm}^{-3}$  or lower to prevent the mobility from being dislocation-limited.<sup>35</sup>

Multiple methods have been proposed and utilized for reducing the dislocation densities in α-Ga<sub>2</sub>O<sub>3</sub> thin films. The primary method has been growing films thicker and allowing the threading dislocations to recombine.<sup>30</sup> For example, by extending the film thickness from 200 nm to 8 μm, Ma *et al.* were able to decrease the dislocation density from  $1 \times 10^{11}$  to  $3 \times 10^9 \text{ cm}^{-2}$ .<sup>33</sup> It has also been demonstrated that for the growth of α-Ga<sub>2</sub>O<sub>3</sub> by mist CVD on *m*-plane sapphire substrates, higher growth rates lead to reduced quality, but a thin template of higher quality α-Ga<sub>2</sub>O<sub>3</sub> grown slowly followed by quickly grown α-Ga<sub>2</sub>O<sub>3</sub> results in the entire film being high quality.<sup>30</sup> This suggests that the quality of the nucleation layer at the substrate may be the most important determinant of the overall film quality.

We believe that the key to improving the transport properties of doped α-Ga<sub>2</sub>O<sub>3</sub> is lowering the density of dislocations that thread into the layer, either by reducing the number of dislocations that form or increasing the rate at which they recombine in the buffer layer so that fewer propagate into the doped layer. In this

paper, we employ a two-step growth process consisting of a relaxed  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layer with  $x = 0.08 \pm 0.02$  grown at high substrate temperature ( $T_{\text{sub}}$ ) to act as a pseudosubstrate. On top of this pseudosubstrate, a Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> overlayer is grown that is strained to the underlying relaxed buffer layer grown at low  $T_{\text{sub}}$  to produce a conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layer. The two-step process improves the quality of the nucleation layer and allows us to achieve record high conductivities and mobilities in  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films. We additionally introduce a second, thicker buffer layer in some  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films to demonstrate that spacing from the relaxation layer can improve the transport properties.

## METHODS

For the growth and doping of  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> films by S-MBE, we use an elemental aluminum (Alfa Aesar, 6N purity) source, a Ga<sub>2</sub>O source formed by mixing liquid gallium (Alfa Aesar, 7N purity) and Ga<sub>2</sub>O<sub>3</sub> powder (Alfa Aesar, 5N purity), and a SiO<sub>2</sub> (Kurt J. Lesker, 4N purity) source. The gallium + Ga<sub>2</sub>O<sub>3</sub> mixture is made with a 0.4 molecular fraction of oxygen, which thermodynamic calculations conclude will provide a 99.98% pure Ga<sub>2</sub>O molecular beam.<sup>36</sup> The Ga<sub>2</sub>O mixture is loaded into a custom BeO (Materion Thermalox<sup>®</sup> CR, 99.7% purity) crucible contained in a Veeco 400 g SUMO<sup>®</sup> source. The limited purity of the BeO crucible for the Ga<sub>2</sub>O source is not a significant concern since the Ga<sub>2</sub>O mixture produces a growth flux at relatively low temperatures.<sup>36</sup> Aluminum is loaded into a pyrolytic boron nitride (p-BN) crucible contained in a Veeco 40 cc dual-filament effusion cell. SiO<sub>2</sub> is loaded into a polycrystalline BeO crucible (Materion Thermalox<sup>®</sup> 995, 99.5% purity) contained in a Veeco 10 cc high-temperature effusion cell. Prior work found that containing SiO<sub>2</sub> in a polycrystalline Al<sub>2</sub>O<sub>3</sub> crucible with 99.8% purity led to significant iron contamination being seen in the secondary-ion mass spectrometry (SIMS) of Si-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> films, which is a compensating defect in Ga<sub>2</sub>O<sub>3</sub>.<sup>37</sup> All of the crucibles are loaded into retractable, differentially pumped MBE effusion cells that are mounted on a Veeco Gen10 MBE system. Implementing a 400 g SUMO cell for Ga<sub>2</sub>O rather than a 40 cc effusion cell for S-MBE is seen to extend the source lifetime and significantly reduces spitting from the source. The smaller opening of the SUMO crucible has led to issues with clogging, but additional tantalum heat shielding and a 100–200 °C tip offset can prevent clogging.

The fluxes of the Ga<sub>2</sub>O and aluminum molecular beams are calibrated *in situ* using a quartz crystal microbalance (QCM). The flux of the SiO<sub>2</sub> beam is calibrated by using an Arrhenius fit of fluxes determined by x-ray reflectivity (XRR) measurements on amorphous SiO films deposited onto sapphire substrates held at room temperature in vacuum with the SiO<sub>2</sub> effusion cell at a high temperature of ~1450 °C in combination with a Hall effect measurement of Si-doped homoepitaxial  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> grown with the SiO<sub>2</sub> effusion cell at a lower temperature of ~1250 °C.<sup>37</sup> The 10 × 10 mm<sup>2</sup>  $m$ -plane Al<sub>2</sub>O<sub>3</sub> substrates are produced by Kyocera and are oriented with an unintentional miscut of <0.1°. Prior to growth, the substrates are cleaned with Micro-90<sup>®</sup>, a DI water rinse, acetone, and then isopropanol in a sonicator and annealed in open air at 1000 °C for 5 h. The  $m$ -plane Al<sub>2</sub>O<sub>3</sub> substrates are back-side coated with a 10 nm thick titanium adhesion layer, followed by 200 nm of platinum to enable uniform radiative thermal heating. For reasons that

are not yet fully understood,  $m$ -plane substrates irregularly have a regular step-and-terrace morphology when examined by atomic force microscopy (AFM), which we call “stepped,” while most substrates are smooth and featureless, identical to their as-received state, which we call “flat.” It has previously been shown that an intentional miscut toward the  $a$ -direction can support step formation.<sup>38</sup> We believe that the origin of the irregular step formation is our use of well-oriented  $m$ -plane substrates with no control over the slight (<0.1°) miscut direction. Using intentionally miscut  $m$ -plane substrates may allow for better control over step formation. Figure S1 of the [supplementary material](#) shows AFM images of substrates that demonstrate typical flat [Fig. S1(a)] and stepped [Fig. S1(b)] morphologies. Films G1–G31 were grown on flat substrates, and films G32–G36 were grown on stepped substrates.

The  $T_{\text{sub}}$  is measured with an optical pyrometer operating at a wavelength of 980 nm. The substrate holders are machined from Haynes<sup>®</sup> 214<sup>®</sup> alloy. The aluminum source only has the base filament heated to prevent metal from climbing over the crucible walls and damaging the source.<sup>39</sup> The use of an elemental aluminum source is also motivated by Ellingham diagrams, which show aluminum to readily reduce Ga<sub>2</sub>O and form Al<sub>2</sub>O, which should readily incorporate into the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> films at the growth conditions used.<sup>40</sup>

The Ga<sub>2</sub>O flux ( $\Gamma_{\text{Ga}_2\text{O}}$ ) was fixed at  $(5.8 \pm 0.8) \times 10^{14}$  molecules cm<sup>-2</sup> s<sup>-1</sup> during the growth of all films for both the unintentionally doped (UID) buffer layers and the Si-doped layers. For the UID  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layers, the aluminum flux ( $\Gamma_{\text{Al}}$ ) was fixed at  $(1.5 \pm 0.5) \times 10^{13}$  atoms cm<sup>-2</sup> s<sup>-1</sup>. Except for films G38–G40, the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layers are  $(37 \pm 2)$  nm thick and the overlying  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films are 200–700 nm thick. The relatively high  $T_{\text{sub}}$  (700–800 °C) provides enough thermal energy for the  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> or  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffers to maintain high crystallinity during relaxation and effectively make a pseudosubstrate. Following the UID buffer, an  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layer was deposited at various  $T_{\text{sub}}$  and background pressures of distilled ozone (~80% O<sub>3</sub> + 20% O<sub>2</sub>,  $P_{80\% \text{O}_3}$ ). An effusion cell containing SiO<sub>2</sub> was used to dope the overlying  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers by supplying an SiO flux.<sup>37,41</sup> The  $T_{\text{sub}}$  for the Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers ranged from 460–550 °C and  $P_{80\% \text{O}_3}$  ranged from  $5 \times 10^{-7}$ – $5 \times 10^{-6}$  Torr. [Table I](#) contains the  $T_{\text{sub}}$ ,  $P_{80\% \text{O}_3}$ , mobile electron carrier concentration measured by Hall effect ( $n$ ), and the transport properties of every film shown or discussed in this study.

X-ray diffraction (XRD) rocking curves,  $\theta$ – $2\theta$  scans, and XRR measurements were employed at room temperature to characterize the phase purity and structural quality of each sample using a PANalytical Empyrean XRD system utilizing copper  $K\alpha_1$  radiation. Rocking curves were measured in a triple-axis configuration using a 220 germanium analyzer crystal. The surfaces of films and substrates were measured by AFM with an Asylum Research Cypher Environmental AFM. The transport properties were determined from Hall effect measurements at 300 K in the van der Pauw geometry using a Nanometrics HL5500 Hall system.

Each sample had indium contacts soldered onto each corner to reduce the contact resistance and enable Hall effect measurements. Select samples had these indium contacts removed with 10 min of soaking in concentrated HCl. These samples then had 10 nm of titanium followed by 110 nm of gold deposited as  $1 \times 1$  mm<sup>2</sup> squares in each corner using electron-beam evaporation with a custom shadow

**TABLE I.** The growth conditions and resulting transport properties of all  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffers and  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films in this study. All buffer layers were grown at a pressure of  $5 \times 10^{-6}$  Torr. The XRD rocking curve (RC) widths are those of the 300 peak for the overlapping buffer layer and  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> film. The listed  $n$ ,  $\mu$ , and  $\sigma$  are the values determined by Hall effect measurements. For electrically insulating samples, we list the expected donor concentration,  $N_D$ .

ID	$T_{\text{sub,Buffer}}$ (°C)	$T_{\text{sub,Film}}$ (°C)	$P_{80\% \text{O}_3, \text{film}}$ (Torr)	300 RC FWHM (arc sec)	$n$ (cm <sup>-3</sup> )	$\sigma$ (S/cm)	$\mu$ (cm <sup>2</sup> /V s)	Contact-type
G1	N/A	480	$5 \times 10^{-6}$	Amorphous	Insulating ( $N_D = 6.6 \times 10^{19}$ )	...	...	In
G2	700	540	$1 \times 10^{-6}$	970	Insulating ( $N_D = 2.3 \times 10^{18}$ )	...	...	In
G3	750	480	$5 \times 10^{-6}$	1020	$4.2 \times 10^{19}$	420	62	In
G4	750	480	$1 \times 10^{-6}$	890	$4.3 \times 10^{19}$	390	55	In
G5	750	480	$5 \times 10^{-6}$	600	$4.2 \times 10^{19}$	540	79	In
G6	750	500	$1 \times 10^{-6}$	990	$2.8 \times 10^{19}$	77	17	Ti/Au
G7	750	525	$1 \times 10^{-6}$	960	Insulating ( $N_D = 1.1 \times 10^{20}$ )	...	...	Ti/Au
G8	750	550	$1 \times 10^{-6}$	1000	Insulating ( $N_{\text{Si}} = 3.9 \times 10^{20}$ )	...	...	Ti/Au
G9	750	460	$5 \times 10^{-6}$	660	$4.8 \times 10^{19}$	570	73	In
G10	750	470	$5 \times 10^{-6}$	720	$4.2 \times 10^{19}$	510	74	In
G11	700	550	$5 \times 10^{-6}$	1020	Insulating ( $N_D = 9.9 \times 10^{18}$ )	...	...	In
G12	750	550	$5 \times 10^{-6}$	870	$1.8 \times 10^{18}$	4.4	15	In
G13	700	550	$1 \times 10^{-6}$	890	Insulating ( $N_D = 1.1 \times 10^{19}$ )	...	...	Ti/Au
G14	700	550	$5 \times 10^{-7}$	830	Insulating ( $N_D = 1.4 \times 10^{19}$ )	...	...	Ti/Au
G15	750	500	$5 \times 10^{-7}$	1010	Insulating ( $N_D = 1.0 \times 10^{19}$ )	...	...	In
G16	750	480	$5 \times 10^{-6}$	670	$4.9 \times 10^{19}$	650	82	In
G17	750	480	$5 \times 10^{-6}$	820	$1.9 \times 10^{19}$	170	54	In
G18	750	480	$5 \times 10^{-6}$	810	$4.2 \times 10^{19}$	320	73	In
G19	750	480	$5 \times 10^{-6}$	800	$5.3 \times 10^{18}$	72	53	In
G20	750	500	$2 \times 10^{-6}$	880	$3.5 \times 10^{18}$	64	68	In
G21	725	550	$5 \times 10^{-6}$	1010	Insulating ( $N_D = 1.5 \times 10^{19}$ )	...	...	In
G22	750	500	$5 \times 10^{-6}$	800	$3.0 \times 10^{19}$	320	67	Ti/Au
G23	750	500	$5 \times 10^{-6}$	760	$2.4 \times 10^{19}$	190	49	Ti/Au
G24	750	500	$5 \times 10^{-6}$	870	$2.0 \times 10^{19}$	200	71	In
G25	750	500	$5 \times 10^{-6}$	830	$1.2 \times 10^{19}$	130	71	Ti/Au
G26	750	500	$5 \times 10^{-6}$	840	$6.4 \times 10^{18}$	67	64	Ti/Au
G27	750	500	$5 \times 10^{-6}$	830	$2.2 \times 10^{18}$	29	79	Ti/Au
G28	750	500	$5 \times 10^{-6}$	810	$9.9 \times 10^{17}$	7.6	51	Ti/Au
G29	750	500	$5 \times 10^{-6}$	830	$4.2 \times 10^{17}$	2.2	31	Ti/Au
G30	750	500	$5 \times 10^{-6}$	820	Insulating ( $N_D = 3.2 \times 10^{17}$ )	...	...	Ti/Au
G31	800	540	$1 \times 10^{-6}$	930	$3.7 \times 10^{18}$	16	26	Ti/Au
G32	730	425	$1 \times 10^{-6}$	1510	$2.0 \times 10^{19}$	57	20	In
G33	730	480	$1 \times 10^{-6}$	1270	$1.1 \times 10^{19}$	67	40	In
G34	800	540	$1 \times 10^{-6}$	1270	$1.1 \times 10^{19}$	140	79	In
G35	800	570	$1 \times 10^{-6}$	1290	$7.9 \times 10^{18}$	14	11	In
G36	800	600	$1 \times 10^{-6}$	1290	Insulating ( $N_D = 3.0 \times 10^{19}$ )	...	...	In
G37	725, 550	500	$2 \times 10^{-6}$	1100	$2.9 \times 10^{19}$	420	90	In

mask made of alloy 316 stainless steel. Samples with Ti/Au contacts and carrier concentrations below  $10^{19}$  cm<sup>-3</sup> had highly Si-doped ( $5 \times 10^{19}$  cm<sup>-3</sup>)  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>  $1 \times 1$  mm<sup>2</sup> triangular contacts regrown onto the corners with S-MBE before the Ti/Au contacts were applied. The highly Si-doped regrown contacts were grown using a sapphire shadow mask for 10 min at  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr and  $T_{\text{sub}} = 490$  °C, resulting in a thickness of ~100 nm. Following the deposition of the Ti/Au contacts, the samples were annealed at 500 °C in a flowing N<sub>2</sub> atmosphere for 2 min. Figure S3 of the

supplementary material shows  $I$ - $V$  curves of sample G27 after multiple annealing temperatures, with 500 °C being the optimal annealing temperature. Our annealing recipe is similar to previous results that found Ti/Au to form ohmic contacts with minimal  $R_c$  on  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> after a 450 °C anneal.<sup>42</sup> Table I lists whether each film had indium contacts or Ti/Au contacts for the Hall effect measurement.

Scanning transmission electron microscopy (STEM) was used to confirm the density of threading dislocations in sample G5.

A cross-section lamella was prepared using the Thermo Fisher Helios G4 UX Focused Ion Beam. Protective amorphous carbon and platinum layers were deposited onto the lamella and prepared with a final milling step of 2 keV to reduce damage. The STEM measurements were taken with an aberration-corrected Thermo Fisher Spectra 300 CFEG operated at 300 keV. The density of threading dislocations was also estimated for film G5 by measuring rocking curves on one symmetric peak and multiple asymmetric peaks and fitting the FWHM as a function of the inclination angle of each reflection. This method has previously been demonstrated to match well with STEM measurements for  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown on *c*-plane sapphire.<sup>33</sup> The Burgers vectors for edge dislocations and screw dislocations of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> on *m*-plane sapphire are taken to be  $b_e = \langle 110 \rangle$  and  $b_s = \frac{1}{3} \langle 1\bar{1}0 \rangle$  for *m*-plane  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>, which have magnitudes of 4.98 and 2.88 Å, respectively.<sup>34</sup>

SIMS measurements were made on samples G6, G8, G16, G23, G25, G26, G29, G30, and G37 using a Hiden Analytical SIMS Plus Workstation. The Hiden system used an O<sub>2</sub><sup>+</sup> ion source as the primary beam to profile the sample with a voltage and current for the analysis of 2 kV and 50 nA, respectively. The O<sub>2</sub><sup>+</sup> primary ion beam was oriented at 45° relative to the sample surface, while a MAXIM quadrupole mass analyzer was angled at 30° to the probe axis. The crater area, scan density, electronic gating, system base pressure, and the pressure during profiling were 500 × 500 μm<sup>2</sup>, 100 × 100 pixel<sup>2</sup>, 10% of the raster area, 5.0 × 10<sup>-10</sup> Torr, and 2.0 × 10<sup>-8</sup> Torr, respectively.

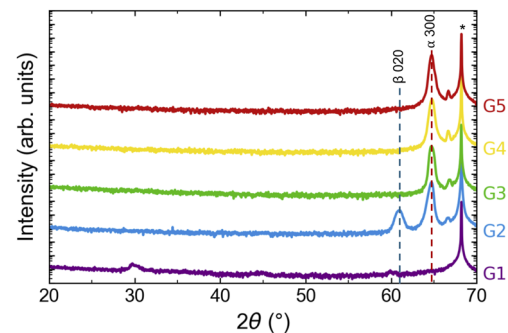
Prior to the SIMS measurements that are shown in this manuscript, two  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films were grown to create SIMS standards. The first of these undoped films consisted of two  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers that were grown at  $T_{\text{sub}} = 725^\circ\text{C}$  and  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr and 550 °C and  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr on flat *m*-plane Al<sub>2</sub>O<sub>3</sub> substrates, with a total thickness of 670 nm. The second undoped film was composed of two  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers that were grown at  $T_{\text{sub}} = 725^\circ\text{C}$  and  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr and 575 °C and  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr on flat *m*-plane Al<sub>2</sub>O<sub>3</sub> substrates, with a total thickness of 480 nm. Both samples were implanted with 10<sup>14</sup> cm<sup>-2</sup> doses of boron, beryllium, silicon, and iron by CuttingEdge Ions<sup>®</sup>. These calibration standards in combination with the depth of the SIMS crater measured by profilometry, allowed the measured SIMS intensities to be converted to elemental concentrations.

The optical bandgaps,  $E_g$ , of films G22–G30 were measured using ellipsometry to confirm phase purity, as  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> has the highest optical bandgap of all Ga<sub>2</sub>O<sub>3</sub> phases. All 16 Mueller elements for the films were collected using a JA Woolam RC2 ellipsometer for azimuthal angles 0° (along the optical axis) and 90° (orthogonal to the optical axis) for photon energies in the range of 0.73–6.42 eV with a resolution of 0.03 eV. The angles of incidence varied from 45° to 75° in 5° steps. An initial anisotropic B-Spline model for the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layer on *m*-plane  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> was developed with a thickness of (37 ± 2) nm determined by XRR and a composition of  $x = 0.08 \pm 0.02$  determined by the 2θ peak position in the  $\theta$ -2θ XRD scan. This film was a model of the buffer that was used in all films G2–G36, unless otherwise specified. The  $\alpha$ -(Al<sub>0.08</sub>Ga<sub>0.92</sub>)<sub>2</sub>O<sub>3</sub> model was used in the subsequent ellipsometry analysis. To further estimate the thickness and roughness of the  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films, an initial anisotropic fit using a Cauchy model in difference mode  $[(n_0(\hbar\omega) - n_e(\hbar\omega))|_{\hbar\omega < 3\text{eV}} = 0.02]$ , where  $n_0$  and  $n_e$  are the ordinary and extraordinary indices of refraction at photon energy  $\hbar\omega$ ,

respectively, was done in the transparent region (<3 eV).<sup>43</sup> These values were fixed for the subsequent analysis. The dielectric function was modeled with two Kramers–Kronig enforced B-Spline layers with a node spacing of 0.03 eV, which were simultaneously fit for the two azimuthal angles. From the dielectric function, we write the corresponding absorption coefficient  $\alpha(\hbar\omega) = \frac{4\pi \text{Im}(\sqrt{\epsilon(\hbar\omega)})}{\lambda}$ , where  $\epsilon$  is the permittivity and  $\lambda$  is the wavelength. The absorption coefficient was then used to extract the Tauc bandgap of the material using a Tauc-plot fit of  $E_g \propto (\hbar\omega\alpha)^2$  for both the ordinary and the extraordinary dielectric functions. We observe that the bandgap along the extraordinary direction is larger than along the ordinary direction. We report the ordinary bandgap from the Tauc fitting as the optical bandgap of the material.

## RESULTS AND DISCUSSION

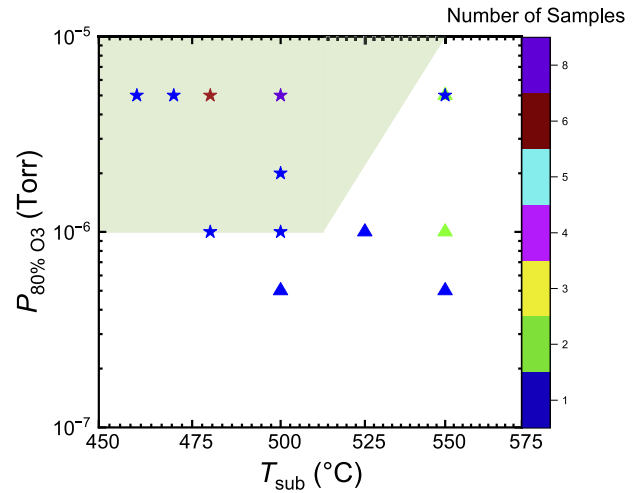
The  $\theta$ -2θ XRD scans of samples G1–G5 are compared in Fig. 1 to determine whether the implementation of a high-temperature relaxed  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> or a relaxed  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layer can template the low-temperature growth of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>. Each sample was grown with different buffer layer conditions to determine which works best. G1 is used as a control sample with no buffer layer; only an Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> film is grown. G2 and G3 have  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> buffer layers, while G4 and G5 have  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffers with  $x = 0.08 \pm 0.02$ . Figure S2 of the supplementary material shows the  $\theta$ -2θ scan of an example  $\alpha$ -(Al<sub>0.08</sub>Ga<sub>0.92</sub>)<sub>2</sub>O<sub>3</sub> buffer layer and confirms that the  $\alpha$ -(Al<sub>0.08</sub>Ga<sub>0.92</sub>)<sub>2</sub>O<sub>3</sub> buffer is relaxed from the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> substrate. The example  $\alpha$ -(Al<sub>0.08</sub>Ga<sub>0.92</sub>)<sub>2</sub>O<sub>3</sub> buffer shown in Fig. S2 was grown for 10 min at  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr and  $T_{\text{sub}} = 750^\circ\text{C}$ . The  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer must have low aluminum content so that the Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layer grown upon it does not relax from the buffer layer, which would introduce additional threading dislocations. G2 and G4 have their Si-doped layers grown at  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr, while G3 and G5 have their Si-doped layers grown at  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr. The growth conditions for all films and buffers, as well as their resulting transport properties, are listed in Table I.



**FIG. 1.**  $\theta$ -2 $\theta$  XRD scans of samples G1–G5 over the  $2\theta = 20^\circ$ – $70^\circ$  range, offset along the vertical intensity axis for clarity. The 300 peak of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> substrate is marked by the \* symbol, and the 300  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> and 020  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> reflections are labeled above the dashed red and dashed blue lines, respectively. The broad, weak peaks at 30° and 44° seen only in G1 are believed to be  $\kappa$ -Ga<sub>2</sub>O<sub>3</sub> impurity phase peaks.

The results in Fig. 1 confirm that the high-temperature buffer is necessary for substantial overgrowth of phase-pure epitaxial  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> since G1 has weak and broad peaks, suggesting limited crystalline growth with low crystalline quality and at least one impurity phase present without a buffer layer. The peaks at 30° and 44° in the  $\theta$ -2 $\theta$  scan of G1 are consistent with  $\kappa$ -Ga<sub>2</sub>O<sub>3</sub> inclusions, and the peak at 60° signals 020  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> formation. With no aluminum in the buffer layer and low  $P_{80\%O_3}$  during doping, sample G2 exhibits no measurable conductivity and shows the formation of the  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> inclusions. In contrast, G3–G5 all show phase-pure epitaxial  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> growth with substantial peak intensity, supporting our hypothesis that a relaxed high-temperature buffer layer could act as a pseudosubstrate and template the growth of phase-pure  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> at low  $T_{\text{sub}}$ . The additional peak at 66.9° in the  $\theta$ -2 $\theta$  scans of G2–G5 is believed to be a hybrid reflection formed by the 200 substrate plus 100 film reflections, which is supported by Fig. S4 of the supplementary material.<sup>44</sup> In Fig. S4(a) of the supplementary material, the 300  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> and the 300  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> peaks appear in both scans with little change in intensity and position, as expected for symmetric peaks. In the  $\Phi = -2.5^\circ$  scan, an extra peak appears at 66.9°, which is within 0.15° of the calculated position for the 200 substrate plus 100 film hybrid reflection,  $2\theta = 67.04^\circ$ . The extreme sensitivity to  $\Phi$  and agreement with the calculated reflection position are consistent with the peak being a hybrid reflection.<sup>44</sup> By changing  $\Phi$  after aligning to the 300  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> substrate peak, we are consistently able to get this 200 substrate plus 100 film hybrid reflection to appear. These peaks appear in all films grown with buffer layers and do not appear in films without buffer layers. Figure S4(b) shows that the film and substrate  $\Phi$  scans have intense reflections at all values of  $\Phi$ , which is consistent with symmetric film reflections, while the suspected hybrid reflection shows four narrow, sharp peaks in  $\Phi$ , which is consistent with hybrid reflections.

Of the samples shown in Fig. 1, only samples G3, G4, and G5 are conductive. In contrast to sample G2, the Si-doped layer of sample G3 was grown at a higher  $P_{80\%O_3}$  and remains phase pure with a conductivity of 422 S/cm. The Si-doped layer of G2 was grown at  $T_{\text{sub}} = 540^\circ\text{C}$  rather than  $T_{\text{sub}} = 480^\circ\text{C}$  at which all the other Si-doped layers of the samples in Fig. 1 were grown, which may also contribute to the lack of conductivity. The difference between G2 and G3 suggests that  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> formation and poor transport may be issues for films grown in low  $P_{80\%O_3}$  and high  $T_{\text{sub}}$  conditions, even with  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> buffer layers. In contrast to G2 and G3, samples G4 and G5 had aluminum added to their buffer layers and both remained phase-pure. Additionally, samples G4 and G5 have relatively high conductivities of 387 S/cm and 544 S/cm, respectively. The FWHM of the 300 rocking curve of the Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layer also decreases from 970 to 890 arc sec with the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer, suggesting that the structural quality improves when aluminum is added to the buffer layer. The addition of aluminum to the buffer also improves transport in high  $P_{80\%O_3}$  conditions, which can be seen by G5 having higher conductivity than G3. The improved structural quality offered by the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer is corroborated as the 300 rocking curve FWHM of the Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layer improves from 1020 arc sec for G3 to 600 arc sec for G5. Comparing G2–G4 and G3–G5, the  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer successfully prevents  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> inclusions and leads to higher conductivity. Additionally, increasing  $P_{80\%O_3}$  to  $5 \times 10^{-6}$  Torr during growth and doping leads to both G3 and G5 being more conductive than G2 and G4.

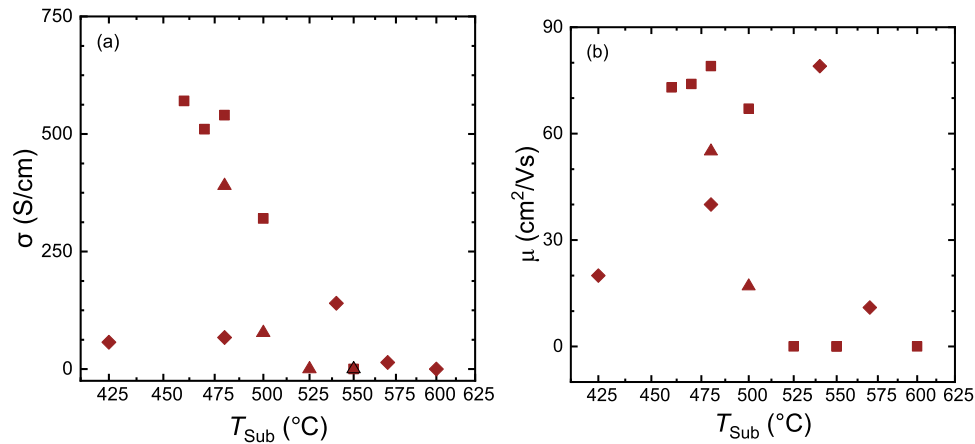


**FIG. 2.** A  $P_{80\%O_3}$  -  $T_{\text{sub}}$  map showing the growth conditions resulting in conductive films, marked by stars, and insulating films, marked by triangles. In the figure, the minimum conductivity for a sample to be considered conductive is 1 S/cm. The color of each point disambiguates how many samples were grown in identical conditions. The light green region marks the growth conditions where all films turned out conductive.

Overall, Fig. 1 confirms that a buffer is necessary for the growth of conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> at low  $T_{\text{sub}}$  and that adding aluminum to the buffer can further improve the structural quality, while higher  $P_{80\%O_3}$  values may improve the transport properties.

Next, to determine which growth conditions are most favorable for growing conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> with S-MBE, samples G3–G28 were grown over the range of  $P_{80\%O_3} = 5 \times 10^{-7}$ – $5 \times 10^{-6}$  Torr and  $T_{\text{sub}} = 460$ – $550^\circ\text{C}$  and are compared in Fig. 2. We see that our  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films are always electrically insulating if grown at a  $P_{80\%O_3} < 1 \times 10^{-6}$  Torr. We similarly find that Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown at  $T_{\text{sub}} > 550^\circ\text{C}$  are never conductive, while Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown at  $500 < T_{\text{sub}} < 550^\circ\text{C}$  are inconsistently conductive but only at relatively high  $P_{80\%O_3}$ . Specifically, only one of the three films grown at  $T_{\text{sub}} = 550^\circ\text{C}$  and  $P_{80\%O_3} = 5 \times 10^{-6}$  Torr is conductive, suggesting that  $T_{\text{sub}} \leq 500^\circ\text{C}$  is still preferable even at high  $P_{80\%O_3}$ . This is believed to be either a result of lower  $P_{80\%O_3}$  and higher  $T_{\text{sub}}$  conditions, which increases the surface lifetime of adatoms and the chance of the donor, Si<sup>4+</sup>, reaching a dislocation and being compensated, or of the donor failing to find an oxygen adatom and incorporating as SiO, thereby behaving as an acceptor.<sup>17,26,30</sup>

The green region of Fig. 2 marks the most optimal conditions for the growth and Si-doping of conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> by S-MBE. For the most ideal growth conditions,  $T_{\text{sub}} \leq 500^\circ\text{C}$  and  $P_{80\%O_3} \geq 1 \times 10^{-6}$  Torr, the growth of conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> is highly reproducible, and all 19 Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films are conductive. G30 is excluded from the figure because it is expected to have  $n \leq 1 \times 10^{17} \text{ cm}^{-3}$ , and forming ohmic contacts remains challenging. G30 is insulating by both 2-point multimeter readings and Hall effect measurements. If this sample was included, 19 of 20 samples in the green region would be considered conductive. The requirement of low  $T_{\text{sub}}$  for conductivity supports the previous finding that



**FIG. 3.** (a) A comparison of the conductivity of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films as a function of the substrate temperature at which the Si-doped layer was grown. (b) A plot of the mobility of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films as a function of  $T_{\text{sub}}$ . The samples with  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr have a target  $n$  of  $(9.0 \pm 2.5) \times 10^{19} \text{ cm}^{-3}$ , while the samples with  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr have a target  $n_e$  of  $(6.20 \pm 0.15) \times 10^{19} \text{ cm}^{-3}$ . The films grown on stepped substrates have a target  $n$  of  $(1.85 \pm 0.25) \times 10^{19} \text{ cm}^{-3}$ . Triangle markers represent samples grown on “flat” substrates with  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr, square markers represent samples grown on “flat” substrates at  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr, and diamond markers represent samples grown on “stepped” substrates.

a relaxed buffer layer with good crystalline quality is necessary for the growth of an overlying conductive doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> film since growth and doping of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> at low  $T_{\text{sub}}$  is only possible with a pseudosubstrate template.<sup>17,26,30</sup>

Next, to find the growth conditions for S-MBE that result in the best transport properties, the conductivity and mobility of samples G4–G11, G22, and G32–G36 are plotted as a function of  $T_{\text{sub}}$  and  $P_{80\% \text{O}_3}$  in Fig. 3. The samples grown at  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr have a target donor concentration of  $N_D = (6.20 \pm 0.15) \times 10^{19} \text{ cm}^{-3}$ , and samples grown at  $P_{80\% \text{O}_3} = 1 \times 10^{-6}$  Torr have target  $N_D = (9.0 \pm 2.5) \times 10^{19} \text{ cm}^{-3}$ . The films grown on stepped substrates have  $N_D = (1.85 \pm 0.25) \times 10^{19} \text{ cm}^{-3}$ . Consistent with the findings of Fig. 2, conductivity is only observed in the films grown at  $T_{\text{sub}} \leq 500^\circ \text{C}$ . The mobility and conductivity are higher for the higher  $P_{80\% \text{O}_3}$  samples at all  $T_{\text{sub}}$  values, again consistent with the findings of Fig. 2. The specific growth conditions and transport properties of each film are given in Table I.

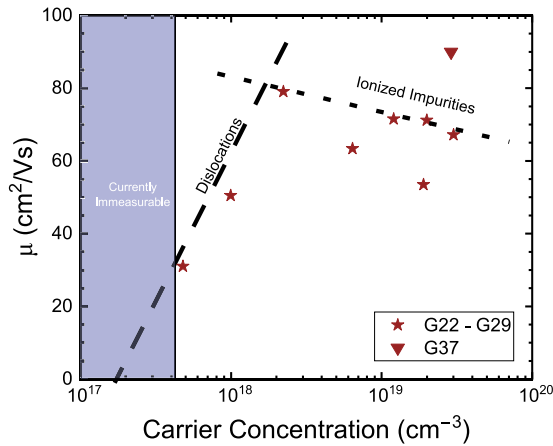
Samples G32–G36 were grown on stepped substrates rather than the flat substrates used throughout the rest of this paper. The comparison in Fig. 3 primarily shows that the growth of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> on stepped substrates is seen to result in reduced  $\sigma$  and  $\mu$  compared to flat substrates at the same mobile carrier concentration,  $n$ , especially at  $T_{\text{sub}} \leq 500^\circ \text{C}$ , where flat substrates produce  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films with the best transport properties. The lower  $\sigma$  values may result from the lower target  $N_D$  for the films on stepped substrates. A lower  $N_D$  is expected to result in higher Hall mobilities, which is not observed across the majority of tested  $T_{\text{sub}}$ . This suggests that the lower  $\sigma$  and  $\mu$  values are due to the reduced crystalline perfection of the  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films.

Our results show that conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> thin films can be grown at  $T_{\text{sub}}$  as low as  $425^\circ \text{C}$  on stepped substrates, which is back-end-of-line (BEOL) compatible. This is in agreement with prior reports that  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> is the widest bandgap semiconductor that

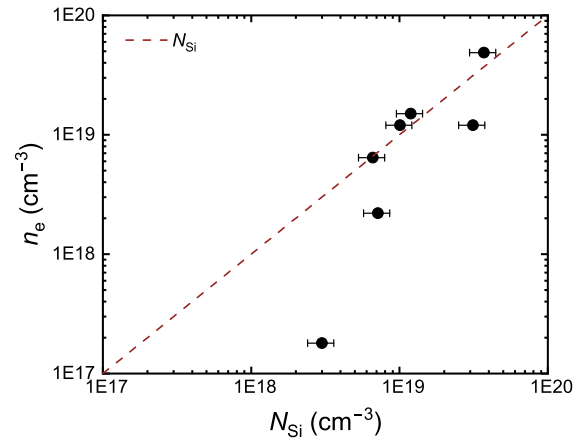
can be deposited in BEOL conditions, but our mobilities under BEOL conditions are nearly 100 times higher.<sup>45,46</sup> This growth in BEOL conditions requires a  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> buffer, which is grown at high  $T_{\text{sub}}$  in S-MBE and cannot be done fully in BEOL conditions. It may be possible to grow the initial relaxed  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> buffer with ALD or mist CVD, both of which have demonstrated growth of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> in BEOL conditions, followed by highly conductive  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> grown by S-MBE.<sup>45,46</sup> Similar conditions have not been tested for flat substrates, but the advantage in transport properties from  $T_{\text{sub}} = 460$ – $500^\circ \text{C}$  suggests that BEOL temperatures may result in better films on flat substrates. One potential advantage offered by the stepped substrates is that exclusively in the temperature range of  $T_{\text{sub}} > 500^\circ \text{C}$ , the mobility is higher than on the flat substrates at the same  $n$ . A possible explanation for this is that the step-and-terrace morphology may enable 2D step-flow growth, which requires increased  $T_{\text{sub}}$ .<sup>47</sup>

With the ideal range of  $P_{80\% \text{O}_3}$  and  $T_{\text{sub}}$  for the growth and doping of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> by S-MBE determined in Figs. 2 and 3, we selected  $P_{80\% \text{O}_3} = 5 \times 10^{-6}$  Torr and  $T_{\text{sub}} = 500^\circ \text{C}$  as the conditions for a doping series. All films in this series have  $\alpha$ -(Al<sub>*x*</sub>Ga<sub>1-*x*</sub>)<sub>2</sub>O<sub>3</sub> buffer layers with  $x = 0.08 \pm 0.02$  that are approximately  $(37 \pm 2)$  nm thick and overlying Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers with thicknesses in the range of  $(415 \pm 55)$  nm. The target  $n_e$  for the films in this series ranged from  $3.2 \times 10^{17}$ – $9.8 \times 10^{19} \text{ cm}^{-3}$ . The resulting mobilities and conductivities are plotted as a function of the mobile carrier concentration determined by Hall effect measurements in Fig. 4. The samples in Fig. 4 are G22–G30, and the growth conditions and resulting transport properties for each film are listed in Table I. The goal of this series was to determine at what carrier concentration dislocation scattering began to dominate, which defines the range of carrier concentrations that can be grown effectively.

In Fig. 4, the peak mobility in our  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films occurs for a carrier concentration of  $3 \times 10^{19} \text{ cm}^{-3}$ , and the transition to



**FIG. 4.** A plot of the mobility and conductivity of films G22–G29 and G37 as a function of their measured carrier concentration. The values for  $\mu$ ,  $\sigma$ , and  $n$  come from Hall effect measurements. The films marked with a star in this plot are all grown with  $P_{80\%O_3} = 5 \times 10^{-6}$  Torr and  $T_{sub} = 500^\circ\text{C}$  as the growth conditions for the Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  overlayers and  $P_{80\%O_3} = 5 \times 10^{-6}$  Torr and  $T_{sub} = 750^\circ\text{C}$  as the growth conditions for the UID  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  buffer layers. The Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  overlayers for the samples marked by stars have thicknesses of  $415 \pm 55$  nm, and the buffer layers are approximately  $(37 \pm 2)$  nm thick. The sample marked with a triangle, G37, has an initial buffer layer grown at  $P_{80\%O_3} = 5 \times 10^{-6}$  Torr and  $T_{sub} = 550^\circ\text{C}$  that is  $\sim 50$  nm thick and an additional UID buffer layer that is grown at  $P_{80\%O_3} = 5 \times 10^{-6}$  Torr and  $T_{sub} = 550^\circ\text{C}$  that is  $\sim 310$  nm thick. The dashed line represents the region of dislocation-limited  $\mu$  as a function of  $n$ , and the dotted line represents the region of ionized-impurity scattering limited  $\mu$  as a function of  $n$ , for samples G22–G29.



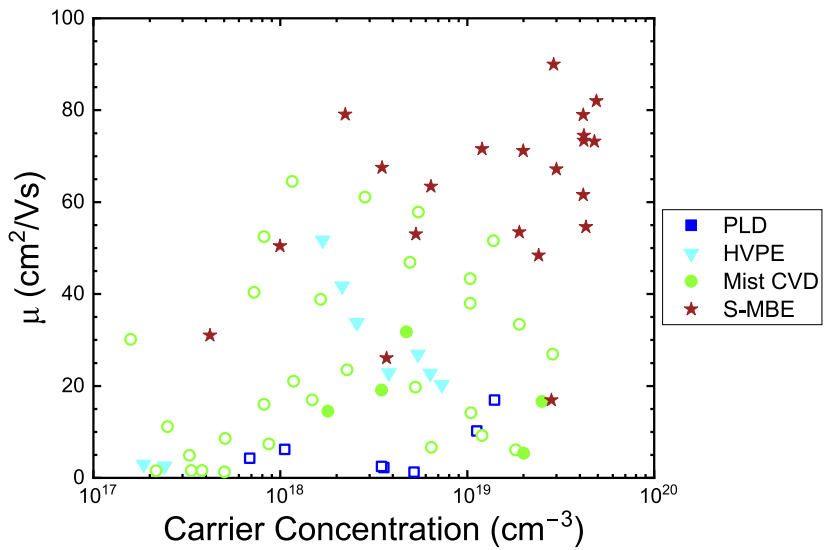
**FIG. 5.** A plot of  $n_e$ , determined by Hall measurements, as a function of the concentration of silicon atoms,  $N_{Si}$ , determined by SIMS measurements. Error bars of  $\pm 20\%$  in the SIMS measured  $N_{Si}$  are included for each data point.<sup>48,49</sup> The dashed line shows the expected trend of  $n_e$  as a function of  $N_{Si}$  if 100% of the incorporated Si donors are electrically active.

dislocation-limited mobility occurs around  $2 \times 10^{18} \text{ cm}^{-3}$ . At high carrier concentrations, the expected decrease in mobility due to increased ionized-impurity scattering occurs. Below a carrier concentration of  $2 \times 10^{18} \text{ cm}^{-3}$ , the mobility plummets as the mobile carrier concentration falls, as the dominant scattering mechanism changes from ionized-impurity scattering to dislocation scattering. This occurs because the concentration of ionized impurities decreases and screens the dislocation scattering less, increasingly allowing dislocation scattering to limit the mobility. For homoepitaxial films of other semiconductors, e.g.,  $\beta\text{-Ga}_2\text{O}_3$ , the dislocation densities are many orders of magnitude lower, so the concentration where dislocations begin to dominate is extremely low. Metastable systems, such as  $\alpha\text{-Ga}_2\text{O}_3$ , however, often require heteroepitaxy since they cannot be stabilized as bulk substrates to enable homoepitaxy. The significant lattice mismatch and strain involved in the heteroepitaxial growth of  $\alpha\text{-Ga}_2\text{O}_3$  on sapphire results in a relatively high concentration of  $10^{10}\text{--}10^{11} \text{ cm}^{-2}$  threading dislocations upon relaxation, causing the mobility to be primarily limited by dislocation scattering at carrier concentrations as high as  $2 \times 10^{18} \text{ cm}^{-3}$ , which is consistent with Fig. 5.<sup>27,32–35</sup>

In Fig. 5, the active carrier concentration,  $n_e$ , is plotted as a function of the SIMS determined donor concentration,  $N_{Si}$ , to extract the activation ratio of donors in films G6, G8, G16, G23, G25, G27, G29, G30, and G37. The fit of  $n_e$  as a function of  $N_{Si}$  is the activation ratio of the incorporated silicon and is plotted against the dashed line in Fig. 5, which represents full incorporation of the

supplied flux,  $N_{Si}$ . The error bars on the measured value of  $N_{Si}$  by SIMS are typical of the technique and account for  $\sim 20\%$  error.<sup>48,49</sup> The SIMS measurement of sample G23 is shown as an example in Fig. S5(a) of the [supplementary material](#). Clear interfaces between the film, buffer, and substrate are seen in the SIMS spectra and show that the silicon concentration is relatively constant throughout the doped layer. The activation rate of silicon donors at  $N_{Si} \geq 10^{19} \text{ cm}^{-3}$  is  $\sim 100\%$ . At lower values of  $N_{Si}$ , below  $10^{18} \text{ cm}^{-3}$ , the effective activation ratio rapidly falls to 0. This is expected to be a result of threading dislocations acting as Read cylinders that remove an approximately constant concentration of carriers.<sup>50,51</sup> This reduces the effective activation of low  $N_{Si}$  samples by removing a larger fraction of the active carriers and results in a minimum  $N_{Si}$  where active carriers can be observed. This leads to a limited range within which the doping can be effectively controlled of  $\sim 10^{18} \leq n_e \leq 10^{20} \text{ cm}^{-3}$ . To broaden the range of effective doping, reducing the threading dislocation density is necessary to limit the number of carriers that are lost.

SIMS was also measured on film G8 to investigate why the films grown outside of the ideal growth conditions found in Fig. 3 are electronically insulating. The SIMS of film G8 is shown in Fig. S6 (b) of the [supplementary material](#). Similar to the SIMS of sample G23, clear interfaces between the film, buffer, and substrate are seen. The Si concentration is also consistent throughout the doped layer at a value of  $N_{Si} = 3.9 \times 10^{19} \text{ cm}^{-3}$ . This shows that even in films that are electrically insulating, such as G8, Si is readily incorporated but is not active. Since the insulating films have rocking curves with similar FWHM to the conductive films, the complete deactivation cannot be fully due to Read cylinders. Several hypotheses could explain this behavior. One is that when incorporated at elevated  $T_{sub}$ , significant concentrations of Si incorporate in the 2+ oxidation state and act as compensating defects.<sup>26</sup> Another is that at elevated  $T_{sub}$ , the silicon dopant has a longer diffusion length and enough thermal energy to diffuse to and get pinned at a dislocation, where it forms a Cottrell atmosphere.<sup>52</sup> Such dopant pinning



**FIG. 6.** A comparison of the mobilities of Si- and Sn-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> grown by PLD (squares),<sup>26</sup> HVPE (triangles),<sup>15</sup> mist CVD (circles),<sup>30</sup> and S-MBE (stars, this work) as a function of mobile carrier concentration. Solid markers represent films grown with silicon doping, and empty markers represent films with tin doping. The Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films in this study have mobilities higher than all other reports at  $n \geq 2.2 \times 10^{18} \text{ cm}^{-3}$ .

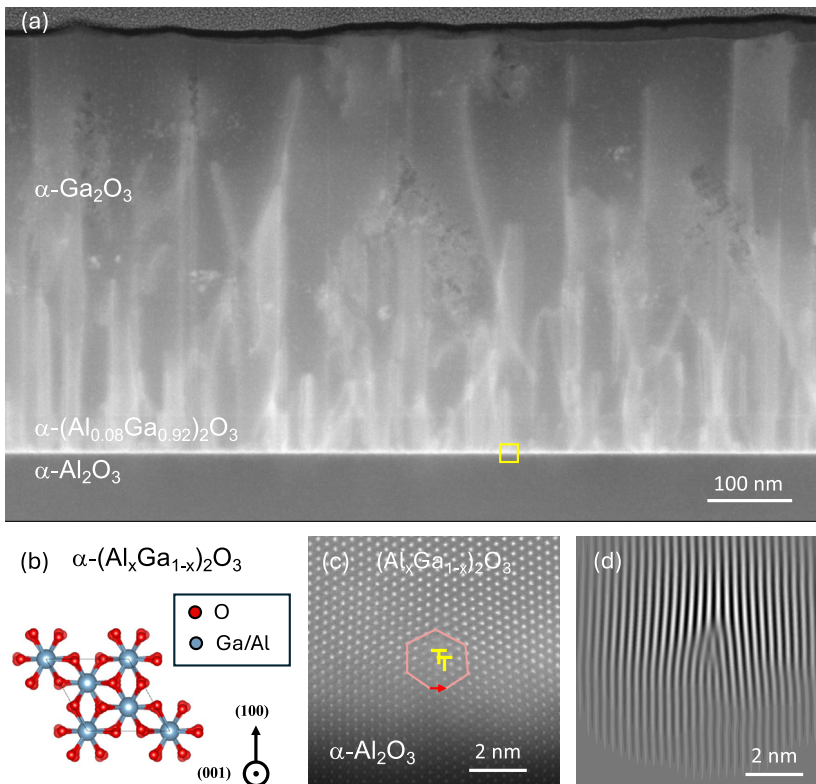
at dislocations (and deactivation because of the percent-level high concentrations commonly observed in Cottrell atmospheres) has been seen in As-doped silicon, B-doped silicon, and Mg-doped GaN.<sup>53–55</sup>

In Fig. 6, we compare the mobilities of our Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films to previous  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> studies in which conducting Sn- or Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films were grown by PLD,<sup>26</sup> HVPE,<sup>14</sup> or mist CVD.<sup>17,18,30</sup> The films grown by S-MBE show a range of active doping, from  $4.2 \times 10^{17}$  to  $4.8 \times 10^{19} \text{ cm}^{-3}$ , reaching the highest mobile carrier concentration of any study to date. Our  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> samples grown by S-MBE with carrier concentrations  $\geq 2.2 \times 10^{18} \text{ cm}^{-3}$  also demonstrate the highest reported mobilities of any study. Twelve of the films shown in this study exceed the previous record of  $\mu = 65 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  set by tin doping with mist CVD.<sup>30</sup> The maximum mobility in this set is  $\mu = 90 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$ , which occurs at a carrier concentration of  $2.9 \times 10^{19} \text{ cm}^{-3}$ . The mobility before the transition to the dislocation-limited regime occurs is  $79 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  at a carrier concentration of  $2.2 \times 10^{18} \text{ cm}^{-3}$ . This transition point is comparable to the transition point for Sn-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> grown by mist CVD.<sup>30</sup> Since the impurity concentrations in mist CVD are high, over  $10^{18} \text{ cm}^{-3}$ , the transition to dislocation scattering is screened by the additional impurities, which suppresses the transition to the regime where the mobility is limited by dislocation scattering.<sup>30</sup> We believe that the record-setting transport properties that we observe are primarily a result of the lower chemical impurity concentrations and donor activation possible with MBE growth. A second reason for the comparably high mobilities could be the superior structural quality of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown by S-MBE. This is supported by an average symmetric rocking curve FWHM of 834 arc sec, which is nearly 20% lower than the previous record of  $\sim 1000$  arc sec for doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> grown on *m*-plane sapphire.<sup>16</sup> As the FWHM of rocking curves increases with the mosaicity of the films, narrower rocking curves are correlated with reduced threading dislocation densities and dislocation scattering.

To better understand the record transport behavior of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films grown by S-MBE, we grew a set of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films at 500 °C on top of  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layers with varied target doping levels ranging from  $3.2 \times 10^{17}$  to  $9.8 \times 10^{19} \text{ cm}^{-3}$ . In Fig. S8 of the [supplementary material](#), the ordinary  $E_g$  of this set of  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films G22–G28 determined by ellipsometry is compared to the measured mobile carrier density,  $n$ , or the target donor concentration,  $N_D$ . All films have bandgaps of 5.36 eV or higher, which confirms the phase purity of the  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> films, as all other phases of Ga<sub>2</sub>O<sub>3</sub> have lower bandgaps. The trend of mobility vs  $n$  for the sample set shows the expected transition from where mobility is limited by ionized impurity scattering to where it is limited by dislocation scattering. The transition between these two scattering-limited mobility regimes is seen to occur at a carrier concentration of  $2.2 \times 10^{18} \text{ cm}^{-3}$ , which is comparable to the value seen in prior studies and suggests similar threading dislocation densities.

To estimate the dislocation density with XRD measurements, we fit a plot of the FWHM of skew-symmetric rocking curves as a function of inclination angle in Fig. S6(b) of the [supplementary material](#).<sup>33,34</sup> This fitting procedure assumes that the out-of-plane tilt broadening comes completely from screw dislocations and the in-plane twist broadening comes entirely from edge dislocations. For the estimation, we measured the rocking curves of the 300, 220, 104, 113, 014, and  $\bar{2}24$  reflections of film G5, which are shown in Fig. S6(a) in the [supplementary material](#). The rocking curves are shown in Fig. S6(a), with their intensities scaled to the same value for clarity. This is the film that is measured with STEM to determine the true density of threading dislocations in Fig. 7. The FWHM for the rocking curves of the 300, 220, 104, 113 014, and  $\bar{2}24$  reflections of film G5 are found to be  $0.167^\circ$ ,  $0.359^\circ$ ,  $0.285^\circ$ ,  $0.264^\circ$ ,  $0.403^\circ$ , and  $0.357^\circ$ , respectively. The inclination angles for the 300, 220, 104, 113, 014, and  $\bar{2}24$  reflections are  $0^\circ$ ,  $30^\circ$ ,  $40.5^\circ$ ,  $51.8^\circ$ ,  $65^\circ$ , and  $72^\circ$ , respectively, for  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub>.

The fit of film G5 in Fig. S6(b) of the [supplementary material](#) gives  $K_{\text{twist}} = 0.39^\circ \pm 0.04^\circ$  and  $K_{\text{tilt}} = 0.20^\circ \pm 0.06^\circ$ . The edge and



**FIG. 7.** (a) A LAADF image of film G5 showing the entire film cross-section along the (001) projection showing a density of threading dislocations of  $(5.6 \pm 0.6) \times 10^{10} \text{ cm}^{-2}$ . The green arrows point to two regions of non-crystalline material. (b) A model of the (001) projection of the  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  structure. The red atoms are the oxygen sublattice, and the blue atoms are the octahedrally coordinated aluminum and gallium atoms. (c) An ADF image of the substrate–buffer interface near a pair of dislocations shows that the aluminum and gallium sublattices of the  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  buffer remain ordered during relaxation. This region is marked by the yellow square in (a). A Burgers circuit is drawn to highlight the direction of the Burgers vector. (d) A Fourier filtered image that exhibits how the threading dislocation cores are generated by masking the  $1\bar{2}10$  fast Fourier transform (FFT) peak of the same region in (c).

screw dislocation densities are estimated by  $D_e = \frac{K_{\text{tilt}}^2}{2\pi \ln(2)b_e^2}$  and  $D_s = \frac{K_{\text{twist}}^2}{2\pi \ln(2)b_s^2}$ , respectively, where  $b_e$  and  $b_s$  are the respective magnitudes of the Burgers vectors of the edge and screw dislocations. The density estimates for our film are  $D_e = 1.1 \times 10^{10} \text{ cm}^{-2}$  and  $D_s = 1.7 \times 10^{10} \text{ cm}^{-2}$ , predicting a total density of threading dislocations of  $D \approx 2.8 \times 10^{10} \text{ cm}^{-2}$ . This density of threading dislocations is around half of typical values reported in the literature for films of similar thicknesses.<sup>33,34</sup> The value is also consistent with the mobility of our films becoming dislocation-limited at a carrier concentration  $2.2 \times 10^{18} \text{ cm}^{-3}$ .<sup>27,31–35</sup>

Film G5 was investigated by low-angle annular dark-field (LAADF-STEM) imaging to assess the density of threading dislocations. Figure 7(a) shows an LAADF image of the film cross-section along the (001) projection. Figure 7(b) is a model of the  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  lattice along the same (001) projection for reference. Figure 7(c) shows an ADF image of a diffuse substrate–buffer interface in which a pair of edge dislocations is present along the interface to accommodate the structural relaxation. Figure 7(d) shows a Fourier filtered image that exhibits the threading dislocation cores generated by masking the  $1\bar{2}10$  fast Fourier transform (FFT) peak.

The STEM imaging confirms that the threading dislocations originate during the structural relaxation in the buffer layer. This supports the idea that optimizing the buffer to reduce threading dislocation density is key to achieving high-quality  $\alpha\text{-Ga}_2\text{O}_3$

growth and improving the transport properties in the future. In Fig. 7(a), the threading dislocation density was measured to be  $(5.6 \pm 0.6) \times 10^{10} \text{ cm}^{-2}$  across the  $\alpha\text{-Ga}_2\text{O}_3$  film. These threading dislocations are part of dislocation half-loops. The segment of these half loops lying at the interface between the  $\alpha\text{-Ga}_2\text{O}_3$  and  $\alpha\text{-Al}_2\text{O}_3$  is a misfit dislocation segment. These segments are needed to relax the lattice mismatch between  $\alpha\text{-Ga}_2\text{O}_3$  and  $\alpha\text{-Al}_2\text{O}_3$ . Ideally, this network of misfit dislocations would be present at only the interface, with no threading segments. The dislocation density was measured at four locations near the surface of  $\alpha\text{-Ga}_2\text{O}_3$ , middle of  $\alpha\text{-Ga}_2\text{O}_3$  film, right above the buffer layer, and within the buffer layer. The measured dislocation density at these four locations was constant within the error of the measurement when the increase in thickness of the STEM lamella wedge was taken into consideration. The reported dislocation density assumes a 25 nm thickness of the lamella wedge at the  $\alpha\text{-Ga}_2\text{O}_3$  surface. This wedge thickness is scaled proportional to the change in intensity from a high-angle annular dark-field (HAADF-STEM) image. This measured density of threading dislocations is relatively low for the film thicknesses compared to other conducting  $\alpha\text{-Ga}_2\text{O}_3$  films on which STEM has been performed and is within approximately a factor of 2 of the XRD-based estimation shown in Fig. S6 of the supplementary material. This shows that the XRD-based estimation may be a useful tool for estimating the threading dislocation density of  $\alpha\text{-Ga}_2\text{O}_3$  thin films grown on  $m$ -plane sapphire in addition to  $c$ -plane sapphire.<sup>33</sup>

The high density of threading dislocations increases the chance that the silicon donors migrate to a dislocation core and deactivate. At the threading dislocation densities seen in the STEM imaging, Read cylinders formed around the threading dislocations can be expected to cause carrier depletion and prevent mobile donors below a critical mobile  $n_e$  of  $\sim 1 \times 10^{18} \text{ cm}^{-3}$ .<sup>56,57</sup> Compensation by traps from 0D defects, such as chemical impurities, are also expected to further impact carrier depletion.<sup>58</sup> A combination of carrier depletion by dislocations and impurities is consistent with no measurable conductivity being observed in films with  $n_e < 4.2 \times 10^{17} \text{ cm}^{-3}$  due to complete deactivation of the donors.

In Fig. 7(a), dark, splotchy regions of non-crystalline material are also visible where the  $\alpha\text{-Ga}_2\text{O}_3$  lattice is disrupted. The green arrows point to two examples of these regions. It is unclear at this point what causes these regions to appear and how much they impact the transport properties. Most non-crystalline regions appear near the surface and along dislocation cores, but some regions occasionally appear far from the surface. We suspect that these regions form after growth since there is no impurity phase signal in the optical bandgap measurements with ellipsometry. The ellipsometry measurements occurred before any additional processing was done to the samples, including the HCl surface treatment. Since amorphous  $\text{Ga}_2\text{O}_3$  has a bandgap of 4.1 eV, we would expect these signals to cause the ellipsometry measurements to read a substantially lower  $E_g$  than that of  $\alpha\text{-Ga}_2\text{O}_3$ . As shown in Fig. S7 of the [supplementary material](#), all ellipsometry measurements on these films show bandgaps of 5.36 eV or higher, which suggests that the regions of amorphous  $\text{Ga}_2\text{O}_3$  form during post-processing of contacts.

In Fig. 7(c), a Burgers circuit is drawn to highlight the direction of the Burgers vector. The Burgers vector represents a single unit cell shift in the [010] direction along the (100) plane, which has dissociated into the two edge dislocations present. The Burgers circuit highlights that the misfit stress is being relieved by the formation of two partial edge dislocations of the  $\langle 110 \rangle$  type. This relaxation of misfit stress by the dissociation into two partial edge dislocations is observed consistently across the substrate–buffer interface and allows for the aluminum and gallium sublattice to remain ordered. Annealing the buffer layers at elevated  $T_{\text{sub}}$  may provide enough thermal energy for threading dislocation segments to annihilate with each other, leading to dislocation half loops with longer misfit dislocation segments at the interface and a reduction in the threading dislocation density. The absence of these threading dislocations in the buffer layer should result in a lower threading dislocation being inherited by the conductive overlayer, which is subsequently grown at lower temperatures, and improve the transport properties further.

## CONCLUSIONS

Previously, even with record structural quality enabled by S-MBE, achieving conductivity in single-layer films remained elusive.<sup>22</sup> We now establish that the implementation of  $\alpha\text{-Ga}_2\text{O}_3$  and preferably  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  as a buffer layer, similar to what has been shown by PLD, provides an effective fix for this issue.<sup>26</sup> This

buffer is deposited at a high temperature and acts as a pseudo-substrate that can template  $\alpha\text{-Ga}_2\text{O}_3$  growth.<sup>26</sup> The addition of aluminum to the buffer to form  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  is effective for suppressing any formation of  $\beta\text{-Ga}_2\text{O}_3$  and improves the structural quality of the epitaxial Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  film. By implementing buffer layers, we show that it is possible to grow conductive Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  at temperatures as low as 425 °C, reaffirming that  $\alpha\text{-Ga}_2\text{O}_3$  is the widest bandgap semiconductor with BEOL-compatible growth conditions. Our results show that  $T_{\text{sub}} \leq 500 \text{ °C}$  and  $P_{\text{O}_3} \geq 1 \times 10^{-6}$  Torr are necessary for the highly reproducible growth of conductive Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  thin films by S-MBE. SIMS measurements confirm that silicon is incorporated in the  $\alpha\text{-Ga}_2\text{O}_3$  thin films both inside and outside of this ideal range of growth conditions but is only electrically active inside the ideal range. The lack of conductivity in films grown at elevated  $T_{\text{sub}}$  is believed to be a result of silicon incorporating as  $\text{Si}^{2+}$  or diffusing to the threading dislocations and forming Cottrell atmospheres.<sup>26,53–55</sup>

Under these optimized growth conditions, mobilities as high as  $90 \frac{\text{cm}^2}{\text{V}\cdot\text{s}}$  and conductivities as high as 646 S/cm at room temperature were achieved. Both are the highest values for  $\alpha\text{-Ga}_2\text{O}_3$  thin films grown by any method to date. Compared to  $\alpha\text{-Ga}_2\text{O}_3$  thin films grown by PLD, HVPE, and mist CVD, our S-MBE grown films have higher mobilities for carrier concentrations of  $n \geq 2.2 \times 10^{18} \text{ cm}^{-3}$ , and the highest mobilities ever demonstrated at any  $n$ . The rocking curves of our Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  thin films also have FWHM that are nearly 20% narrower than the narrowest values reported for Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  on  $m$ -plane sapphire.

Cross-sectional STEM on one  $\alpha\text{-Ga}_2\text{O}_3$  film shows that  $(5.6 \pm 0.6) \times 10^{10} \text{ cm}^{-2}$  threading dislocations were generated during the structural relaxation of the buffer layer and reach the surface of the Si-doped  $\alpha\text{-Ga}_2\text{O}_3$  overlayer. We also confirm that a combination of symmetric and asymmetric rocking curves can give a reasonable estimate (within approximately a factor of two) for the dislocation density in  $\alpha\text{-Ga}_2\text{O}_3$  thin films grown on  $m$ -plane sapphire. The density of threading dislocations is relatively low for the film thicknesses of conducting  $\alpha\text{-Ga}_2\text{O}_3$  films but is unlikely to fully explain the improved transport properties. We instead suspect that high activation ratios and improved chemical purity available with S-MBE growth are the primary reasons for the higher mobilities in our films.

The main challenge to further improve transport behavior in  $\alpha\text{-Ga}_2\text{O}_3$  thin films, especially for  $N_D \leq 10^{18} \text{ cm}^{-3}$ , is finding a way to reduce the threading dislocation densities in the doped  $\alpha\text{-Ga}_2\text{O}_3$  film. This is significant because it could allow  $\alpha\text{-Ga}_2\text{O}_3$  to achieve a BFOM multiple times the maximum possible for doped  $\beta\text{-Ga}_2\text{O}_3$ . We believe that optimizing the growth conditions of the  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  buffer layer is the key to reducing dislocation densities in  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  films and maximizing transport properties. The simplest method is growing thicker buffer layers in the same conditions to give the threading dislocations a greater opportunity to terminate with one another. A second option is the use of compositionally graded  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  buffer layers. Another option, which we see as most favorable, is altering the growth conditions of the buffer layers by *in situ* annealing the buffers to lower their threading dislocation densities and create superior  $\alpha\text{-(Al}_x\text{Ga}_{1-x})_2\text{O}_3$  pseudosubstrates.

## SUPPLEMENTARY MATERIALS

See the [supplementary material](#) for additional figures and data about the substrates,  $\alpha$ -(Al<sub>x</sub>Ga<sub>1-x</sub>)<sub>2</sub>O<sub>3</sub> buffer layers, Ti/Au contacts, and Si-doped  $\alpha$ -Ga<sub>2</sub>O<sub>3</sub> layers.

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## AUTHOR DECLARATIONS

### Conflict of Interest

The author D.G.S. has been granted U.S. Patent No. 11,462,402 (4 October 2022) with the title “Suboxide Molecular-Beam Epitaxy and Related Structures.”

## Author Contributions

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## DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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