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Growth of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ by molecular beam epitaxy

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ABSTRACT

The $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ alloy system is a potential new dielectric for compound semiconductors such as GaAs. Using molecular beam epitaxy under metal-modulated growth conditions, we grew the binary oxide, Sm_2O_3 , at two substrate temperatures (100 and 500 °C) and optimized the structural, morphological, and electrical properties of the films. Decreasing the Sm cell temperature suppressed the formation of the monoclinic phase and promoted the growth of the cubic phase. Next, the ternary oxide, $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$, was deposited to investigate the effects of Ga incorporation. Optimization experiments were used to determine the effects of substrate temperature and samarium cell temperature (i.e., growth rate) on film stoichiometry, phase distribution, and microstructure in these films. Films grown at 500 °C showed significant surface roughness and the presence of multiple crystalline phases. Since all of the Sm-based oxides (i.e., samarium oxide with and without gallium) were found to have unbonded Sm metal, annealing experiments were carried out in oxygen and forming gas to determine the effects of annealing on film stoichiometry. The motivation behind annealing in forming gas was to see whether this commonly used technique for reducing interface densities could improve the film quality. GaAs metal-oxide-semiconductor diodes with $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ showed breakdown fields at 1 mA/cm² of 4.35 MV/cm, which decreased with increasing Sm unbonded metal content in the films.

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I. INTRODUCTION

There remains significant interest in advanced compound semiconductor metal oxide semiconductor field effect transistors (MOSFETs) and the related development of gate dielectrics to reduce leakage current, passivate surface traps, and provide electrical isolation between devices.^{1–14} The use of InGaAs, InAs, GaSb, or GaAs as channel materials for MOSFETs with high speed and low power due to their higher electron mobility are hampered by the lack of a stable high-quality native oxide.^{15–27} Typically, these III-V MOS devices cannot reach their full potential because of poor oxide/semiconductor interfacial properties that pin the Fermi-level at the semiconductor surface.^{2,4,5,10} Despite of all the advantages these have as channel materials for MOSFET technology compared to Si, there is still an urgent need for a technology capable of forming insulating layers on them with dielectric and interface properties comparable to the Si-SiO₂ system.^{12–14} Specifically when using high-k gate dielectrics, Coulomb scattering and interface-roughness scattering significantly reduce the carrier mobility. A suitable gate dielectric material must satisfy two requirements:^{4,22–25} (1) the interface between the

semiconductor and the gate dielectric must have a low interface state density (D_{it}) to prevent the Fermi level from being pinned; and (2) the gate dielectric must have a high breakdown field to allow a gate voltage to be established. Samarium oxide (Sm_2O_3) and samarium gallium oxide $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ are potential candidate dielectric materials for III-V MOSFET technology.^{28–32} Sm_2O_3 has a high κ (14–16), high breakdown electric field (5–7 MV/cm), large bandgap (4.33 eV),^{28–32} large conduction offset with GaAs,³³ and low hygroscopic characteristics.^{29,30} Sm_2O_3 has a bixbyite crystal structure that is composed of two interpenetrating face-centered cubic. The lattice parameter of Sm_2O_3 is 5.32 nm representing a lattice mismatch of 3.3% with the zinc blende cubic structure of GaAs (lattice parameter 5.65 nm).

In this work, growth of thin (20–50 nm) samarium oxide and samarium gallium oxide layers on GaAs substrates via plasma-assisted molecular beam epitaxy (MBE) was performed using two growth temperatures (100 and 500 °C) in an attempt to affect the crystallinity (amorphous versus crystalline) of the oxide and samarium cell temperatures that were typically 570 °C. There

was residual Sm metal in the films, which decreased with decreasing Sm cell temperature, but was relatively independent of substrate temperature. The stoichiometry of the oxides was found to be independent of substrate temperature but increased in oxygen to metal ratio as the Sm cell temperature was decreased, as low as 510 °C. Decreasing the Sm cell temperature also suppressed the formation of the monoclinic phase and promoted the growth of the cubic phase. Films grown at higher (500 °C) temperature showed the presence of a crystalline interface but relatively high surface roughness and the presence of multiple crystalline phases. MOS diodes showed breakdown fields at 1 mA/cm² up to 4.35 MV/cm, which was found to decrease with increasing Sm free metal content in the films. Postgrowth annealing under an oxygen plasma of films grown at higher temperatures resulted in an increase in the oxygen to metal ratio but no change in the crystalline phase distribution.

II. EXPERIMENT

A Riber 2300 MBE system was used for all oxide growths.² The growth chamber was evacuated to $1\text{--}5 \times 10^{-9}$ Torr using an Oxford Cryo-Torr 8 cryopump. Pure Sm and Ga were used as metal sources. The temperature of the Knudsen cells was controlled by a FICS 10 controller that adjusts the power output of an external power supply. An MDP21 radio frequency (rf) source from Oxford Applied Research was used as the oxygen source and operated at a frequency of 13.56 MHz with a forward power of 300 W and a reflected power of 2–3 W. Oxygen (99.995%) was supplied to the plasma head using a high purity O₂ mass flow controller (MFC) that had a 3 SCCM full scale range. The substrate temperature was measured using a backside thermocouple, while the GaAs substrate surfaces were cleaned by *ex situ* etching with a solution of H₃PO₄:H₂O (3:80) for 90 s³⁴ and then an *in situ* exposure time of 20–30 min with a hydrogen plasma under vacuum.

The films were characterized by x-ray photoelectron spectroscopy (XPS) using a Perkin-Elmer PHI 5100 ESCA system, Auger electron spectroscopy (AES) using a Perkin Elmer 6600 system fitted with an Ar sputter gun allowing compositional depth profiling, x-ray diffraction (XRD) performed using a Phillips APD 3720 diffractometer with a copper (Cu) x-ray source, atomic force microscopy (AFM) measurements done using a Dimension 3100 system, and reflection high-energy electron diffraction (RHEED) using a Staib electron gun operating at 15 kV. Following oxide deposition, MOS capacitors were fabricated using a standard shadow mask process. All electrical measurements were conducted with a Signatone S-1160 Series General Purpose Analytical Probe Station. Current-voltage (I-V) measurements were performed with an Agilent 4155C Semiconductor Parameter Analyzer. Capacitance-voltage (C-V) measurements were performed on an Agilent 4284A 20 Hz–1 MHz Precision LCR meter connected to a Lab View-based PC. Breakdown voltages were extracted from the I-V plot at a current density of 1 mA/cm². The extracted voltages were then divided by dielectric film thickness to determine the forward and reverse breakdown voltages. The data from the C-V curve were used to determine the interface state density, flat band voltage shift, and dielectric constant. The D_{it} value and capacitance-voltage behavior of MOS diodes consisting of a bilayer structure of Pt (20 nm)/Au (80 nm) contacts on the dielectric were used to

evaluate the relationship between growth conditions and interfacial quality.^{35–38} Preliminary work on developing SmO₂ has been published previously.^{33,38}

III. RESULTS AND DISCUSSION

The initial aim was optimization of growth conditions and their effects on stoichiometry, phase distribution, and microstructure. Since the surface properties of the semiconductor will largely affect the properties of the deposited film, initial experiments focused on deposition of the binary oxide, Sm₂O₃, at two substrate temperatures, followed by growth of the ternary oxide, (Sm_xGa_{1-x})₂O₃, to investigate the effects of Ga incorporation.

A. Growth at low substrate temperature

The data from Secs. III B and III C are shown in the supplemental file S to avoid making the main article overly long⁴⁰ for a summary of XRD, RMS surface roughness, and XPS of film composition. XRD measurements of Sm₂O₃ grown on GaAs at a substrate temperature of 100 °C showed an amorphous nature of the film verified by RHEED measurements, with peaks at 32.15° and 66.53° associated with GaAs (200) and GaAs (400) of the cubic phase (Fig. S1).⁴⁰ XRD scans of films grown at 100 °C showed no peaks related to the oxide film. The RMS roughness value was measured at 1.4 nm (Fig. S2).⁴⁰ XPS measurements confirmed the presence of Sm free metal in the film, with a broad peak located at 1082.5 eV representing the Sm free metal peak and a more dominant peak associated with Sm bonded to O (Fig. S3).⁴⁰ The breakdown field strength was measured at 3.96 MV/cm (Fig. S4).⁴⁰

B. Growth at high substrate temperature

XRD measurements of Sm₂O₃ grown on GaAs at a substrate temperature of 500 °C showed a crystalline nature of the film verified by RHEED measurements. The RHEED images remained streaky for the duration of growth (Fig. S5).⁴⁰ XRD measurements showed a broad peak at 29.95° attributed to Sm₂O₃ (321) of the cubic phase and a peak at 44.15° associated with Sm₂O₃ (403) of the monoclinic phase (Fig. S6).⁴⁰ There was also a small shoulder at 28.33° associated with Sm₂O₃ (222) of the cubic phase. Peaks at 32.15° and 66.53° are associated with GaAs (200) and GaAs (400) of the cubic phase. AFM measurements revealed a smooth film surface with a RMS roughness value of 0.631 nm (Fig. S7).⁴⁰ XPS measurements confirmed the presence of Sm free metal in the film. The small broad peak located at 1078.9 eV represents the Sm free metal peak and the peak at 1082.3 eV represent Sm bonded to O (Fig. S8).⁴⁰ The breakdown field strength was measured at 3.38 MV/cm (Fig. S9).⁴⁰ The C-V characteristics from diodes fabricated on these films showed a very small change in capacitance when swept from depletion to accumulation.

C. Growth and characterization of ternary oxides grown at 100 °C

The degree of crystallinity was investigated to determine a set of growth conditions for growing a thermally and environmentally stable oxide film. A low growth temperature was chosen to promote the growth of an amorphous or a fine-grained poly-crystalline dielectric film that from the previous work has been shown to enhance I-V

performance.^{2,39} A low growth temperature is also ideally suited for these experiments due to the delicate nature of the clean GaAs surface. After the substrate was cleaned, the sample was rotated out of the growth position. The H₂ plasma was discharged, the O₂ plasma was ignited, and the sample was then rotated back into the growth position. The samarium cell shutter and the O₂ plasma shutter were opened simultaneously to initiate oxide growth. This growth procedure was also used for the oxides grown at 300 and 500 °C.

Powder XRD measurements of films grown at 100 °C with a T_{Sm} of 550 °C revealed no evidence of peaks associated with crystalline oxides, suggesting that the films are amorphous or very fine grained polycrystalline. Peaks located at 31.9° and 66.3° are associated with GaAs (200) and GaAs (400). The peak at 59.1° could not be identified with JCPDS files, but it consistently shows up in scans of the GaAs substrate. This peak could be attributed to an artifact due to the polychromaticity of the x-ray source used in the powder system. The growth rate was 12.9 Å/s. The RHEED pattern immediately disappeared at the onset of oxide growth and never returns, also suggesting an amorphous film. TEM micrographs confirmed the presence of an amorphous interfacial region of approximately 65 Å (Fig. 1). The surface was smooth with an RMS roughness measured by AFM of 0.37 nm. An AES survey scan of the oxide surface revealed Sm, Ga, and O concentrations of 45.1%, 4.0%, and 50.9%, respectively. A depth profile showed that the Ga concentration remained constant throughout the thickness of the film, except for a sharp increase in the O and Ga concentration due to the presence of gallium oxide at the interface [Fig. 2(a)]. The cycle interval was 0.20 min/cycle. To confirm that the Ga was being incorporated from the source beam and not from the GaAs substrate, a depth

profile was conducted on a silicon substrate mounted on the same molybdenum block during the same growth run. This sample also showed the presence of Ga throughout the deposited film.

Films grown at 100 °C showed a breakdown field strength as high as 3.63 MV/cm. The C-V characteristics, as shown in Fig. 2(b), clearly showed accumulation in the forward direction, but full depletion in the reverse direction was not reached before reverse breakdown occurred. The gate voltage was swept from depletion to accumulation.

To investigate the effect of Sm cell temperature, films were grown using Sm cell temperatures ranging from 510 to 570 °C. As expected, the growth rate increased with increasing cell temperature, Fig. 3(a). AES indicated that the Ga composition ranges from 1.3% to just over 10%. XPS of the deposited films showed evidence of residual Sm metal in the films, which was highest at the highest Sm

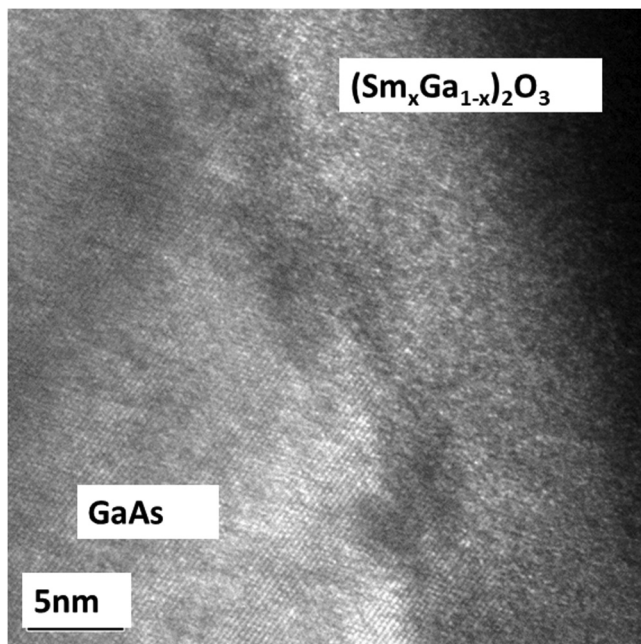


FIG. 1. TEM micrograph of (Sm_xGa_{1-x})₂O₃ grown at a substrate temperature of 100 °C and a Sm cell temperature of 550 °C.

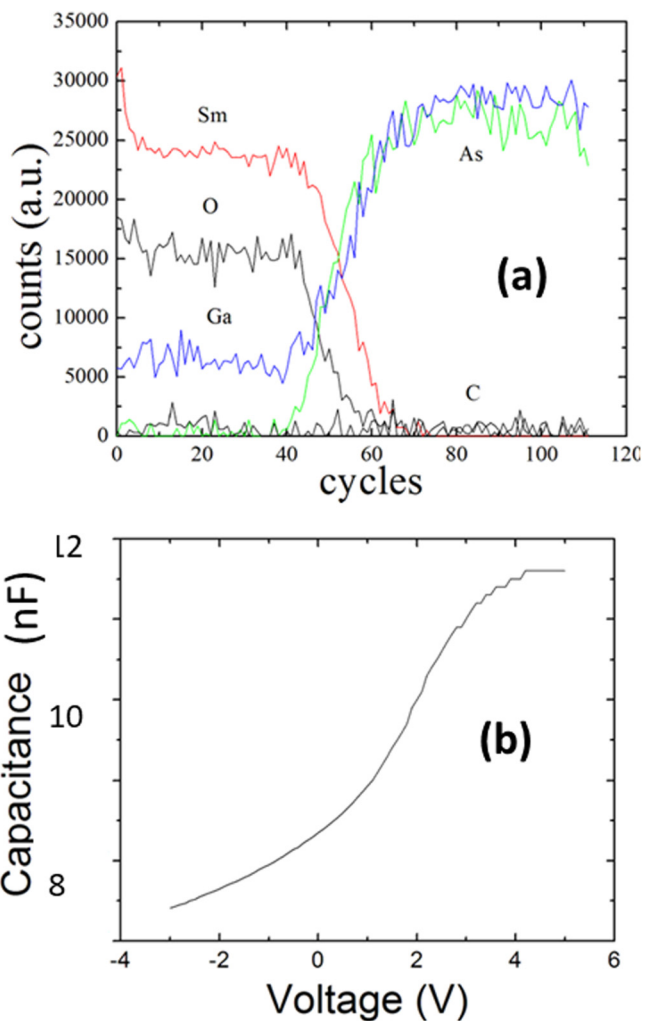


FIG. 2. (a) Depth profile of (Sm_xGa_{1-x})₂O₃ grown at 100 °C and a Sm cell temperature of 550 °C obtained with AES. (b) Capacitance-voltage of (Sm_xGa_{1-x})₂O₃ grown at a substrate temperature of 100 °C and a Sm cell temperature of 550 °C. The frequency was 1 MHz.

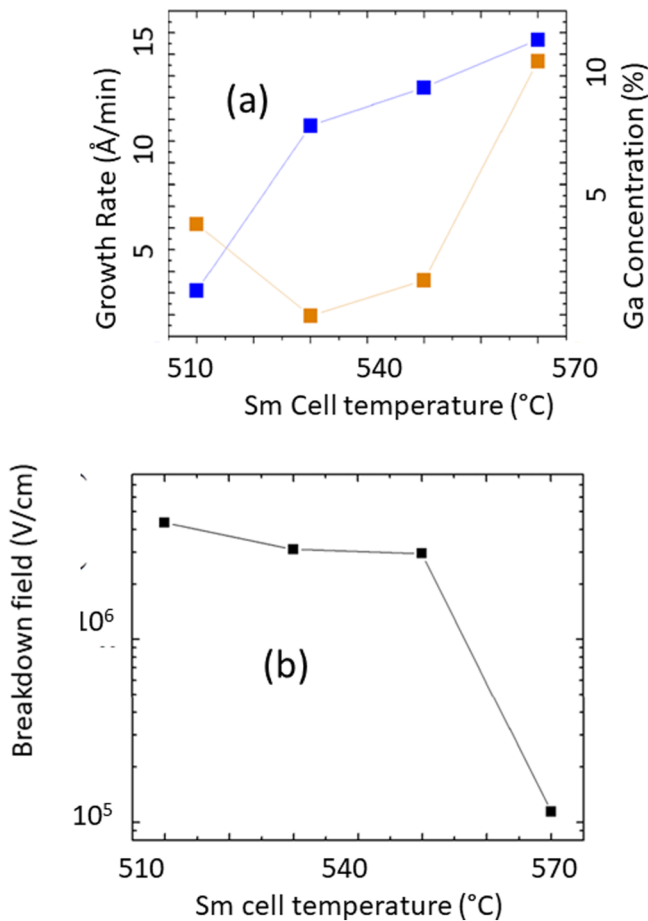


FIG. 3. (a) Growth rate (blue squares, uppermost plot) and Ga concentration (orange squares, lower plot) determined by AES in $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 100 °C as a function of Sm cell temperature. (b) Breakdown voltages for diodes with $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ layers grown at a substrate temperature of 100 °C as a function of Sm cell temperature.

cell temperature (Table I). The AFM root mean square roughness is also given in the table and is accurate to two significant figures. The reason for the presence of residual metal is most likely due to the reduced oxygen to metal ratio present during growth given that the Sm flux is increasing with increasing cell temperature. As one would expect, the breakdown field was lowest in the film with the largest amount of free metal, as shown in Fig. 3(b). The layer grown at $T_{\text{Sm}} = 550$ °C showed the highest oxygen to metal ratio, the lowest RMS surface roughness (Fig. 4), and had a good breakdown field. Subsequent growths were performed using this cell temperature.

D. Growth and characterization of ternary oxides grown at higher substrate temperatures

Films grown at 300 °C using the same procedure as in Sec. III C showed streaky-dashed RHEED patterns, which appeared

TABLE I. List of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ films grown with different Sm cell temperatures showing the oxide, metal, and oxygen XPS peak intensities, ratio of XPS peak intensities, and breakdown field. The substrate temperature was 100 °C for all samples.

Sm cell temp (°C)	Oxide peak counts	Sm metal peak counts	O peak counts	Ratio metal-oxidized metal counts	Breakdown field (MV/cm)
510	42 011	6830	9150	0.16	4.35
530	41 386	7373	8161	0.18	3.11
550	30 467	5635	6904	0.19	3.63
570	23 907	9599	5759	0.40	0.11

around 5 min into the oxide growth corresponding to thickness of ~5.4 nm (i.e., growth rate of 1.08 nm/s). The initial streaky RHEED pattern of the GaAs substrate disappeared after 1 min of oxide growth. After 30 min of oxide growth, the RHEED pattern showed a mixture of arcs and lines suggesting the polycrystalline nature of the film at the end of growth. The background pressure in the chamber was 5×10^{-6} Torr. Powder XRD measurements revealed a textured morphology, as shown in Fig. 5(a). Peaks at 28.9°, 42.2°, and 52.3° are associated with the cubic phase of Sm_2O_3 (222), Sm_2O_3 (134), and Sm_2O_3 (026) orientations, respectively. Peaks located at 47.3° and 54.8° are associated with the monoclinic phase of Sm_2O_3 (602) and Sm_2O_3 (514) orientations. The remaining

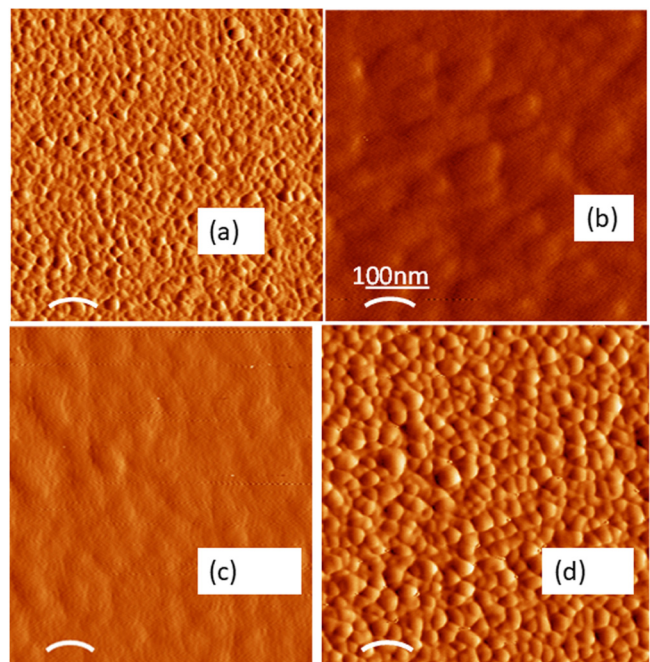


FIG. 4. AFM images of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ layer grown at a substrate temperature of 100 °C and a Sm cell temperature of 510 (a), 530 (b), 550 (c), and 570 °C (d).

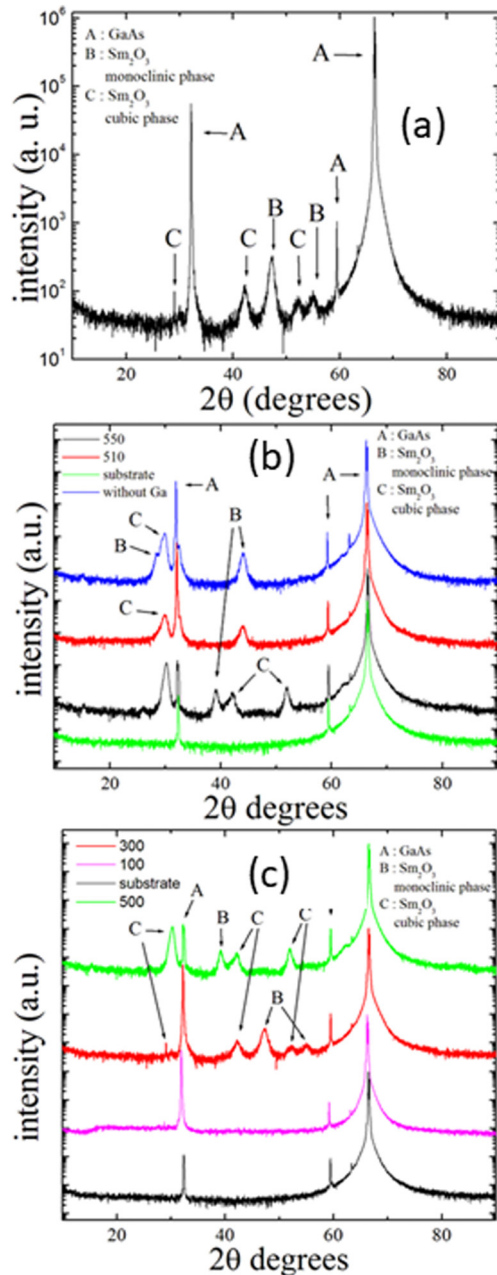


FIG. 5. (a) XRD scan for $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 300 °C and a Sm cell temperature of 550 °C. (b) XRD scans of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at Sm cell temperatures of 510 °C (second from top, red curve) and 550 °C (third from top, black curve) at a substrate temperature of 500 °C. The GaAs substrate scan is shown for comparison (bottom green curve), while the top blue curve is that for growth without Ga. (c) XRD scans of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at 500 °C (top green curve), 300 °C (second from top, red curve), and 100 °C (second from bottom magenta curve) substrate temperatures, respectively (beginning from the topmost curve). The Sm cell temperature was 550 °C for all films. XRD scan of a GaAs substrate is shown for comparison (bottom black curve).

peaks in the spectrum are attributed to the GaAs substrate. An Auger survey scan confirmed Sm, Ga, and O concentrations of 49.7%, 6.5%, and 43.9%, respectively. Gallium was also detected throughout the thickness of the oxide with a sharp increase in Ga concentration occurring around 25 cycles into the depth profile with a cycle interval of 0.20 min/cycle. The XPS analysis of the films showed an oxygen to Sm metal ratio roughly comparable to that obtained in films grown at 100 °C (Table II). The film showed an RMS roughness of 0.7 nm. Electrical characterization of diodes fabricated from the material showed a breakdown field strength of 3.68 MV/cm, and poor C-V behavior with no indication of accumulation or complete depletion before breakdown occurred.

Films grown at 500 °C showed a bright streaky RHEED pattern with thick diffuse lines after 12 s of oxide growth. As the growth proceeds, the lines became faint and arcs began to appear. Sm cell temperatures of 510 and 550 °C were investigated. The XRD analysis indicated that the 550 °C cell temperature enhanced the formation of the cubic phase [Figs. 5(b) and 5(c)]. Therefore, a 550 °C cell temperature was used for subsequent growths.

As compared to the films grown at 300 °C with the same Sm cell temperature, the crystal quality of the cubic phase was enhanced based on an increase in intensity of the peak located at 30.1° [Fig. 5(c)]. This peak is labeled C and shows up only on the XRD scan of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 500 °C and a Sm cell temperature of 550 °C [green curve in Fig. 5(c)]. Auger survey scans show the concentrations of Sm, Ga, and O to be 54.2%, 3.1%, and 42.7%, respectively. The Ga and O concentration show a sharp increase at 55 cycles of the depth profile with a cycle interval of 0.20 min/cycle [Fig. 6(a)]. The XPS analysis of the films showed an O to Sm metal ratio roughly comparable to that obtained in films grown at 100 °C (Table II). The RMS roughness value was 1.0 nm, representing a significant increase with substrate temperature. The TEM micrographs clearly show evidence of atomic registry at a crystalline interface [Fig. 6(b)]. The breakdown field strength was 2.95 MV/cm, and the C-V characteristics indicated a flat band voltage shift of 1 V accumulation in the forward direction and depletion in the reverse direction. Since the C-V curves with this sample showed a significant (≥ 3 pF) change in capacitance as the device was swept from depletion to accumulation, hysteretic C-V curves were also collected. Measurements at 10 kHz clearly showed the hysteretic behavior of the C-V curves. The gate voltage was swept from depletion to accumulation and back to depletion.

E. Growth and characterization of bilayer $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ oxide stack

A dielectric layer with high breakdown field is desirable from an electrical standpoint allowing for the growth of thinner oxides with less leakage.^{2,39} Polycrystalline oxide films deposited at low substrate temperatures typically produce better breakdown behavior than more textured or single crystal films deposited at higher substrate temperatures. Conversely, the crystalline interfaces obtained at higher substrate temperatures typically produce lower interface state densities.

One method of combining the best characteristics of both of these microstructures is to grow the interfacial oxide layer at a high

TABLE II. $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ films grown with different substrate temperatures showing oxide, metal, and oxygen XPS peak intensities, ratio of XPS peak intensities, breakdown field, and RMS roughness.

T_s (°C)	Intensity of oxide peak (counts)	Intensity of Sm metal peak (counts)	Intensity of oxygen peak (counts)	Ratio of peak intensities (metal: oxidized metal)	Ratio of peak intensities (O:Sm metal)	Breakdown field (MV/cm)	RMS (nm)
100	30 467	5635	6904	0.19	0.19	3.63	0.4
300	38 376	6490	8606	0.17	0.19	3.68	0.7
500	25	5905	7532	0.18	0.19	2.95	1.0

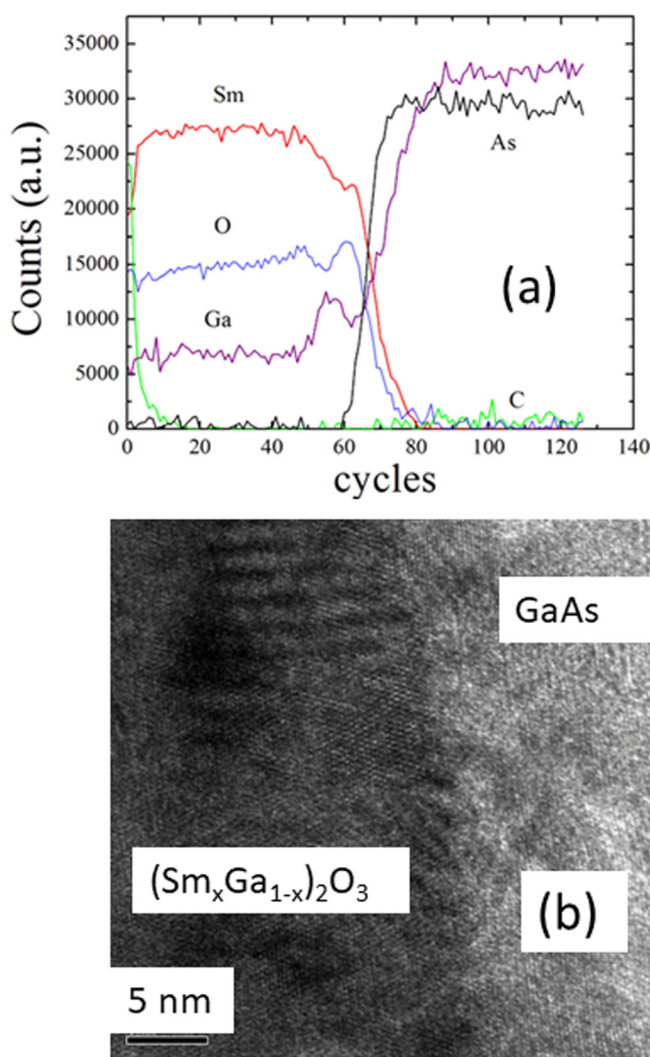


FIG. 6. (a) Auger data showing spike in Ga concentration at the interface of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 500 °C and a Sm cell temperature of 550 °C. (b) Microstructural data of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 500 °C and a Sm cell temperature of 550 °C.

temperature and then drop the temperature for the remainder of the layer. Such a bilayer oxide stack was grown to combine the interfacial properties of oxide growth at 500 °C and the high breakdown field and surface morphology properties of oxide growth at 100 °C. The oxide film grown at 100 °C exhibited an amorphous nature with breakdown field strength of 3.63 MV/cm and an rms value of 0.4 nm. Films grown at 500 °C were crystalline at the interface with a breakdown field strength of 2.95 MV/cm. A flat band voltage shift of 1 V for the oxide film grown at 500 °C is an indication of smaller fixed oxide charge, and the C-V characteristics clearly showed accumulation and depletion.

The growth procedure used for growth of each layer was identical, the only difference being the substrate temperature. The growth of the bilayer oxide stack began with a substrate temperature of 500 °C. The substrate temperature remained at 500 °C for 7.5 min. The substrate was rotated away from the sources and cooled to 100 °C. The sample was then rotated back into the growth position for the remainder of the growth run. The powder XRD measurements of the bi-layer stack showed the presence of cubic phase at 30.01° associated with Sm_2O_3 (321) orientation. The remaining peaks in the spectrum were identified as GaAs peaks. The surface morphology was relatively smooth with an RMS roughness value of 0.5 nm. The breakdown field was measured at 3.13 MV/cm [Fig. 7(a)]. The C-V characteristics of the bilayer oxide were extremely noisy, and thus, a D_{it} measurement could not be determined.

F. Annealing study of Sm-based oxides

Annealing in oxygen was attempted to oxidize residual Sm metal in the oxide layer revealed by XPS measurements. Sm_2O_3 and $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ samples were annealed at 500 °C for 30 min while being exposed to the oxygen plasma. The ratio of intensities of the oxidized metal peak to the Sm free metal peak increased while the ratio of the intensities of the Sm free metal peak to the oxidized metal peak decreased as a result of postgrowth annealing in oxygen of Sm_2O_3 films (Table III). XRD measurements were taken from the sample grown at a substrate temperature of 500 °C before and after annealing at 500 °C for 30 min under an O plasma. The crystal quality degraded as a result of annealing with no change in phase distribution. After postgrowth annealing in O, the RMS roughness value increased from 1.4 to 1.6 nm and 0.6 to 0.7 nm for Sm_2O_3 on GaAs grown at substrate temperatures of 100

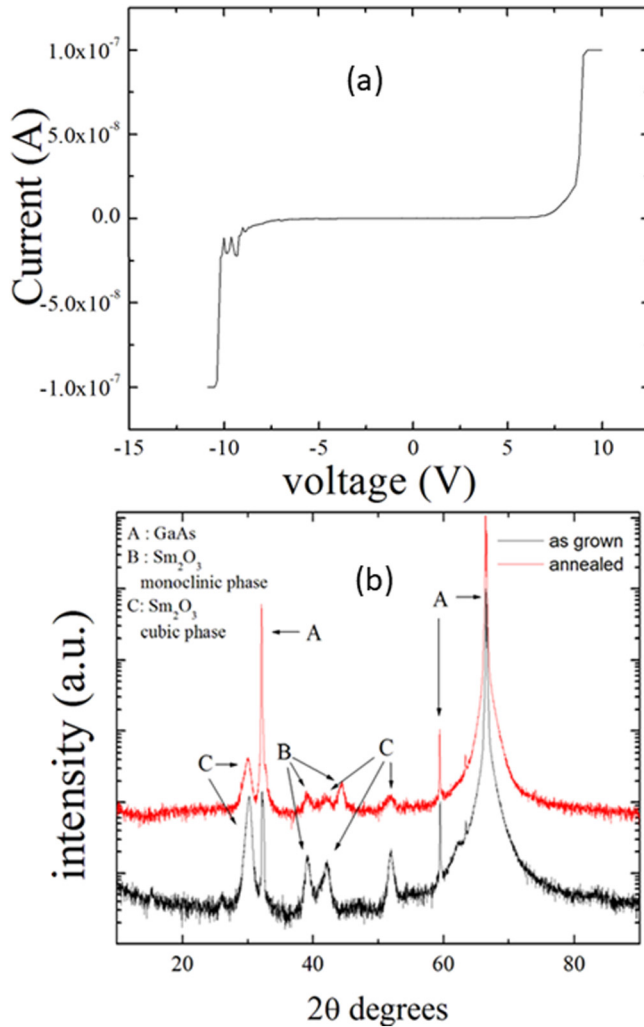


FIG. 7. (a) C-V measurements for a bilayer $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ oxide stack. The oxide was grown at a substrate temperature of 500 °C for the first 7.5 min of growth and at 100 °C for the remaining 22.5 min of growth. (b) Comparison of XRD scans of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ on GaAs at a substrate temperature of 500 °C and a Sm cell temperature of 550 °C before and after annealing under an O_2 plasma for 30 min at 500 °C. The as-grown plots are at bottom, the annealed at top.

and 500 °C, respectively. The sharp peak around 28° is an artifact due to the polychromatic nature of the x-ray source. The annealing under an oxygen plasma in the MBE system did not significantly change either the D_{it} or the breakdown voltage.

The postgrowth annealing of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ films resulted in an increase in the oxygen to metal ratio but effected no change in the crystalline phase distribution [Fig. 7(b) and Table S1].⁴⁰ The breakdown field was increased from 2.95 to 3.37 MV/cm, which is consistent with the increase in the oxygen to metal ratio and it seems to indicate a more insulating film as a result of postgrowth annealing. XRD measurements were taken from a sample grown at

TABLE III. XPS peak intensities and their ratios for a Sm_2O_3 film grown at a substrate temperature of 100 and 500 °C and a Sm cell temperature of 550 °C before and after annealing in oxygen.

T_S (°C)	Ratio of peak intensities			
	(metal; oxidized metal) before annealing	(oxidized metal: oxygen) before annealing	(metal: oxidized metal) after annealing	(oxidized metal: oxygen) after annealing
100	0.296	1.643	0.198	4.08
200	0.191	2.088	0.128	4.62

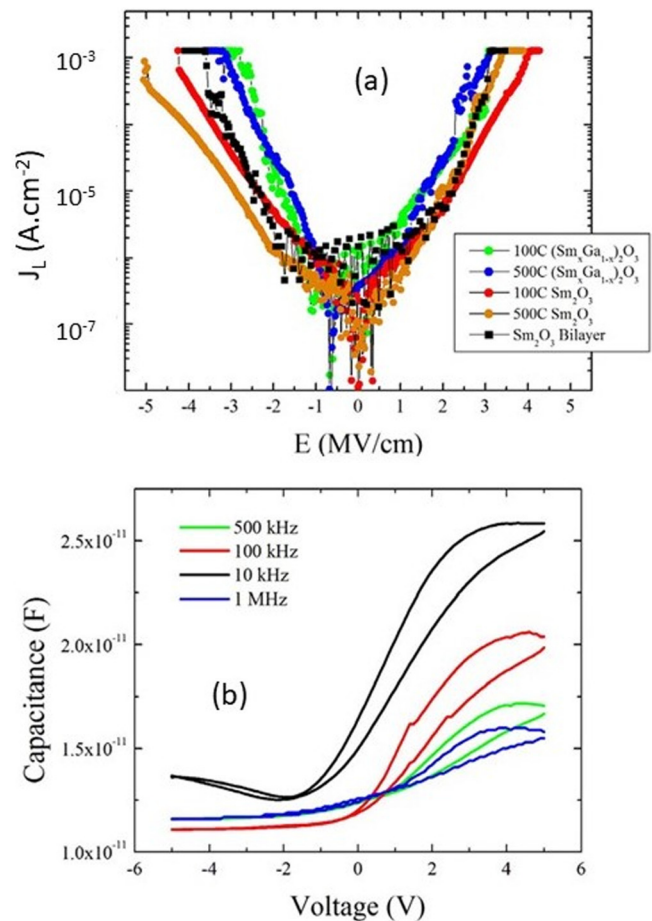


FIG. 8. (a) Comparison of the leakage current vs the breakdown field for Sm_2O_3 bilayer oxide, Sm_2O_3 , and $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ at substrate temperatures of 100 and 500 °C. The ternary films have higher leakage, and this decreases with growth temperature. The bilayer is in between the ternary and binary film results. (b) Family of C-V curves at different frequencies for $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ grown at a substrate temperature of 500 °C and a Sm cell temperature of 550 °C. The capacitance increases with decreasing measurement frequency.

a substrate temperature of 500 °C and a Sm cell temperature of 550 °C before and after annealing at 500 °C for 30 min under an O plasma, as shown in Fig. 7(b). The phase distribution was not affected, but the crystal quality of the film was degraded as a result of postgrowth annealing in oxygen. The RMS roughness value increased from 1.0 to 1.5 nm for $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ on GaAs grown at a substrate temperature of 500 °C.

We also conducted rapid thermal annealing (RTA) of $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ films under forming gas at 300 °C for 45 s. Annealing in a reducing atmosphere should create more dangling bonds by reacting with the oxygen and should result in an increase in the metal to oxygen ratio (Table S2).⁴⁰ The metal to oxygen ratio increased from 0.161 to 0.249. The hysteretic C-V characteristics taken at 10 kHz was measured and compared to the hysteretic C-V curve taken at 10 kHz before annealing in forming gas. Since it was determined that annealing in forming gas results in a film with more unbonded metal, Sm_2O_3 films were not annealed in forming gas.

Postgrowth annealing under an oxygen plasma in the MBE system was successful in oxidizing some of the unbonded Sm metal. However, it was not successful in changing the phase composition from a mixture of monoclinic and cubic oxide to one of the pure cubic phases. Annealing in an RTA under forming gas did not improve the electrical properties of the material and in fact increased the amount of unbonded Sm metal in the oxide. The leakage current as a function of the breakdown field was compared for films grown under all growth conditions [Fig. 8(a)]. It was determined that the binary oxide grown at 100 °C exhibited the best electrical properties in terms of breakdown field. Ternary oxides grown at 500 °C showed the best CV behavior, as displayed in Fig. 8(b).

IV. CONCLUSIONS

Sm_2O_3 metal-modulated growth at a high temperature (500 °C) produced single crystal films, while growth at low temperature (100 °C) yielded amorphous films. $\text{Sm}_2\text{O}_3/\text{GaAs}$ diodes show good breakdown fields (3.9 MV/cm) with relatively low D_{it} . Growth of the ternary $(\text{Sm}_x\text{Ga}_{1-x})_2\text{O}_3$ at 500 °C produced polycrystalline films with a crystalline interfacial layer of approximately 65 Å. Some evidence of Ga segregation at the interface was observed with AES depth profiling. Similar growth at 100 °C produced films that were amorphous. A significant amount of Sm free metal was also observed in all of these films. Despite this, breakdown fields up to 4.35 MV/cm were achieved. Higher growth temperatures produced better CV behavior. Overall, however, the best leakage currents were obtained in pure Sm_2O_3 . Future work could include growth of a digital alloy, which has proven successful for growth of $(\text{Sc}_x\text{Ga}_{1-x})_2\text{O}_3$ alloys.³⁹

Future work on this material system should also focus on further reducing the unbonded metal in the oxide, which should lead to improved electrical behavior. In its present form, the oxide should be adequate for use as a field plate but needs further optimization to be useful as a gate dielectric.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Anthony D. Stewart: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal). **Brent P. Gila:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Project administration (equal); Writing – original draft (equal). **Cammy R. Abernathy:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Writing – original draft (equal). **S. J. Pearton:** Data curation (equal); Investigation (equal); Methodology (equal); Writing – original draft (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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