AFRL-AFOSR-VA-TR-2023-0231

Stannate Perovskite Semiconductor Materials for Novel High Power Devices

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12/21/2022
Final Technical Report

Air Force Research Laboratory
Air Force Office of Scientific Research
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

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1. REPORT DATE 20221221	**2. REPORT TYPE** Final	**3. DATES COVERED** **START DATE** 20190515	**END DATE** 20220914

4. TITLE AND SUBTITLE
Stannate Perovskite Semiconductor Materials for Novel High Power Devices

5a. CONTRACT NUMBER	**5b. GRANT NUMBER** FA9550-19-1-0245	**5c. PROGRAM ELEMENT NUMBER** 61102F
5d. PROJECT NUMBER	**5e. TASK NUMBER**	**5f. WORK UNIT NUMBER**

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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Office of Scientific Research
875 N. Randolph St. Room 3112
Arlington, VA 22203

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/AFOSR RTA1

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-AFOSR-VA-TR-2023-0231

12. DISTRIBUTION/AVAILABILITY STATEMENT
A Distribution Unlimited: PB Public Release

13. SUPPLEMENTARY NOTES

14. ABSTRACT

The overall goal of this project was to analyze the fundamental material properties of the stannate-based perovskite oxide material system, and demonstrate devices that utilize the advantages of this unique class of materials. The project had 3 main components: (1) synthesis of stannate perovskites using hybrid molecular beam epitaxy, (2) analysis to study their fundamental electrical properties, and (3) fabrication and demonstration of devices using these materials. Several other studies of growth and devices on related materials were also performed. The project resulted in over 20 peer-reviewed publications, as well as numerous contributed and invited conference presentations, invited talks and colloquia, which reported numerous breakthroughs in materials science and device performance. A total of nine students were partially or fully supported through this project, with several earning their Ph.D. during the award period.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**
a. REPORT U	**b. ABSTRACT** U	**c. THIS PAGE** U	UU	7

19a. NAME OF RESPONSIBLE PERSON KENNETH GORETTA	**19b. PHONE NUMBER** *(Include area code)* 426-7349

Final Report for Award No. FA9550-19-1-0245
Stannate Perovskite Semiconductor Materials for Novel High Power Devices

For period: 5/15/19 – 9/14/22

Steven J. Koester and Bharat Jalan

Overview

As outlined in the proposal, the overall goal of this project was to analyze the fundamental material properties of the stannate-based perovskite oxide material system, and demonstrate devices that utilize the advantages of this unique class of materials. The project had 3 main components: (1) synthesis of stannate perovskites using hybrid molecular beam epitaxy, (2) analysis to study their fundamental electrical properties, and (3) fabrication and demonstration of devices using these materials. In the early stages, the materials growth was emphasized, and in the later stages, the project concentrated on device fabrication and evaluation of the material electronic properties. In addition, numerous other studies of growth and devices on related materials were also performed.

Summary of Accomplishments

Progress during the award period has been substantial. This award resulted in 21 peer-reviewed publications, numerous invited talks/colloquia and a several prestigious awards. Progress has been accomplished in the three major areas listed above, as well as additional work on related materials systems and devices. The main accomplishments are listed below.

Stannate material synthesis

- We performed a quantitative analysis of native point-defect concentrations at the ppm level in undoped $BaSnO_3$ thin films [4].
- We discovered the existence of a two-dimensional metallic state at the SnO_2-terminated surface of a 1% La-doped $BaSnO_3$ thin film. The observed surface state was shown to have a smaller effective mass ($0.12m_0$) in comparison with the corresponding bulk values, opening the possibility for improved electrical conductivity in $BaSnO_3$ thin films [17].
- Discovery of a new line defect with metallic characteristics in optically transparent $BaSnO_3$ perovskite thin films was reported. The results show that metallic line defects can act as electron sinks, thus affecting dopant activation in lightly-doped films [14].
- An investigation of chemical doping, and solubility limit in a fully-coherent $SrSnO_3$ films was performed [19].
- A comprehensive study of the effect of strain relaxation in the epitaxial, tetragonal phase of Nd-doped $SrSnO_3$ films grown on $GdScO_3$ (110) (GSO) substrates and how it influences the electronic transport properties was performed. This study showed that the tetragonal phase remained fully coherent until it completely transformed into the orthorhombic phase, coinciding with a significant increase in mobility with film thickness [16].
- We made the first-ever demonstration of modulation doping approach in stannates, towards the development of room-temperature high-mobility two-dimensional heterostructures [22].
- A novel self-assembled periodic nanostructure utilizing martensitic phase transformation was discovered. This work showed that $SrSnO_3$ can form reconfigurable periodic nanostructures consisting of regularly spaced regions of sharply contrasted dielectric properties [15].

- An invited review paper was published on the MBE growth of wide bandgap complex oxides [23].

Fundamental materials electronic properties of stannates

- The electron-electron interaction effects in La-doped $SrSnO_3$ films were studied [20].
- We performed a study of magnetism and electrical transport in transparent $BaSnO_3$ films [21].
- We performed a detailed transport study, combining experiment and theoretical calculations, to reveal the intrinsic limiting mechanism for electron mobility in $SrSnO_3$ films [11]. A hysteretic magnetoresistance effect in $SrSnO_3$ films via thermal coupling to dynamic substrate behavior was also discovered [8].
- A comprehensive study of self-heating effects in $SrSnO_3$ thin films was completed. A thermal resistance of 304 $\underline{+}$ 42 K-mm/W was obtained for a 26-nm-thick n-doped $SrSnO_3$ film. For a dissipated power of 0.5 W/mm, the channel temperature rose to 173 °C. This work is important for understanding techniques to minimize self-heating in perovskite-based power devices [1].

Stannate-based electronic devices

- Temperature-dependent analysis and optimization of low-resistance ohmic contacts to n-type-doped $SrSnO_3$ films was performed. It was found that Ti has contact resistance, R_C (specific contact resistivity, ρ_C) as low as 2.4 ± 0.3 Ω-mm (0.03 mΩ-cm^2). It was also found that the R_C (ρ_C) of Ti to the bi-layer films was also ~1 (2) order(s) of magnitude lower than on single-layer controls. Temperature-dependent analysis was also used to extract the barrier height and doping effect for annealed Ti contacts [6].
- We made the first-ever demonstration of a perovskite oxide field-effect transistor with gain at GHz frequencies. RF measurements were performance 0.5-μm gate-length $SrSnO_3$ MESFETs and these devices displayed cutoff frequency, f_T, of 1.31 GHz and a maximum oscillation frequency, f_{max}, of 3.25 GHz at a drain-source voltage, V_{DS}, of 3 V [7].
- High-performance gate-recessed $SrSnO_3$ metal-semiconductor field effect transistors (MESFET) were demonstrated. This work showed that devices using a bi-layer structure, with heavily-doped cap, had roughly 2× better performance than devices fabricated on single-layer channels. The best devices had on-state current as high as 133 mA/mm and extrinsic transconductance of 73 mS/mm. [18].

Related materials studies

- The use of β-$(Al_xGa_{1-x})_2O_3$ capping layer to achieve increased breakdown voltage, V_{BR}, and V_{BR}^2/R_{ON} figure of merit in β-Ga_2O_3 Schottky diodes was evaluated. We demonstrated that the addition of a 30-nm-thick β-$(Al_{0.22}Ga_{0.78})_2O_3$ cap to an n-type β-Ga_2O_3 layer grown by metal organic chemical vapor deposition increases the breakdown voltage by over 50%. This work is a step toward devices that can achieve the full theoretical capabilities of β-Ga_2O_3 [2].
- We investigated the donor state energy in Si-doped β-Ga_2O_3 films grown using metal-organic vapor phase epitaxy. Used temperature-dependent Hall measurements indicated two donor state energies of 33.7 and 45.6 meV, where one is associated with the donor energy of Si, while the other latter suggests a residual donor state [13].
- We investigated the electrostatic doping and transport in β-Ga_2O_3 films, and the results revealed record-high electron mobility of 201 cm^2/Vs, the highest reported value to-date. The

study also revealed novel ways to further increase room-temperature mobility in β-Ga_2O_3 using plasmon-phonon coupling [3].

- A novel solid source metal-organic MBE approach was established for synthesis of "stubborn" metal oxides, where "stubborn" refers to elements which are difficult to evaporate and oxidize [9]. We also demonstrated the growth of nearly-perfect RuO_2 films with the lowest residual resistivity reported to-date [12].
- We reported the first detailed study of defect-electronic structure relationship in $SrTiO_3$ films using x-ray photoelectron spectroscopy [5].

Patents

No invention disclosures or patents were filed related to this project.

Publications

2022 publications

[1] P. Golani, C. N. Saha, P. P. Sundaram, F. Liu, T. K. Truttmann, V. R. S. K. Chaganti, B. Jalan, U. Singisetti, and S. J. Koester, "Self-heating in ultra-wide bandgap n-type $SrSnO_3$ thin films," *Appl. Phys. Lett.* **121**, 162102 (2022).

[2] P. P. Sundaram, F. Alema, A. Osinsky, and S. J. Koester, "β-$(Al_xGa_{1-x})_2O_3/Ga_2O_3$ heterostructure Schottky diodes for improved V_{BR}^2/R_{ON}," *J. Vac. Sci. Technol. A* **40**, 043211 (2022).

[3] A. K. Rajapitamahuni, A. K. Manjeshwar, A. Kumar, A. Datta, P. Ranga, L. R. Thoutam, S. Krishnamoorthy, U. Singisetti, and B. Jalan, "Plasmon-phonon coupling in electrostatically gated β-Ga_2O_3 films with mobility exceeding 200 $cm^2V^{-1}s^{-1}$," *ACS Nano* **16**, 8812-8819 (2022).

[4] K. S. Belthle, U. N. Gries, M. P. Mueller, D. Kemp, A. Prakash, M.-A. Rose, J. M. Börgers, B. Jalan, F. Gunkel, and R. A. De Souza, "Quantitative determination of native point-defect concentrations at the ppm level in un-doped $BaSnO_3$ thin films," *Adv. Func. Mater*. **32**, 2113023 (2022).

[5] D. Lee, F. Liu, T. K. Truttmann, S. A. Chambers, and B. Jalan, "Stoichiometry-dependent surface electronic structure of $SrTiO_3$ films grown by hybrid molecular beam epitaxy," *Appl. Phys. Lett*. **120**, 121604 (2022).

2021 publications

[6] V. R. S. K. Chaganti, P. Golani, T. Truttmann, F. Liu, B. Jalan, and S. J. Koester, "Optimizing Ohmic contacts to Nd-doped n-type $SrSnO_3$," *Appl. Phys. Lett*. **118**, 142104 (2021).

[7] J. Wen, V. R. S. K. Chaganti, T. K. Truttmann, F. Liu, B. Jalan, and S. J. Koester, "$SrSnO_3$ metal-semiconductor field-effect transistor with GHz operation," *IEEE Elect. Dev. Lett*. **42**, 74-77 (2021).

[8] L. R. Thoutam, T. K. Truttmann, A. K. Rajapitamahuni, and B. Jalan, "Hysteretic magnetoresistance in a non-magnetic $SrSnO_3$ film via thermal coupling to dynamic substrate behavior," *Nano Lett.* **21**, 10006-10011 (2021).

[9] W. Nunn, A. K. Manjeshwar, J. Yue, A. Rajapitamahuni, T. K. Truttmann, and B. Jalan, "Novel synthesis approach for "stubborn" metals and metal oxides," *PNAS* **118**, e2105713118 (2021).

[10] W. Nunn, T. K. Truttmann and B. Jalan, “A review of molecular beam epitaxy of wide bandgap complex oxide semiconductors,” *J. Mater. Res.* **36**, 4846-4864 (2021).

[11] T. K. Truttmann, J.-J. Zhou, I.-T. Lu, A. K. Rajapitamahuni, F. Liu, T. Mates, M. Bernardi, and B. Jalan, “Combined experimental-theoretical study of electron mobility-limiting mechanisms in SrSnO$_3$,” *Commun. Phys*. **4**, 241 (2021).

[12] W. Nunn, S. Nair, H. Yun, A. K. Manjeshwar, A. Rajapitamahuni, D. Lee, K. Andre Mkhoyan, and B. Jalan, “Solid-source metal–organic molecular beam epitaxy of epitaxial RuO$_2$,” *APL Mater*. **9**, 091112 (2021).

[13] A. K. Rajapitamahuni, L. R. Thoutam, P. Ranga, S. Krishnamoorthy, and B. Jalan, “Impurity band conduction in Si-doped β-Ga$_2$O$_3$ films,” *Appl. Phys. Lett*. **118**, 072105 (2021).

[14] H. Yun, M. Topsakal, A. Prakash, B. Jalan, J. S. Jeong, T. Birol, and K. A. Mkhoyan, “Metallic line defect in wide-bandgap transparent perovskite BaSnO$_3$,” *Sci. Adv*. **7**, eabd4449 (2021).

[15] A. Prakash, T. Wang, A. Bucsek, T. K. Truttmann, A. Fali, M. Cotrufo, H. Yun, J.-Woo Kim, P. J. Ryan, K. Andre Mkhoyan, A. Alù, Y. Abate, R. D. James, and B. Jalan, “Self-assembled periodic nanostructures using martensitic phase transformations,” *Nano Lett*. **21**, 1246-1252 (2021).

[16] T. K. Truttmann, F. Liu, J. Garcia-Barriocanal, R. D. James, and B. Jalan, “Strain relaxation via phase transformation in high-mobility SrSnO$_3$ films,” *ACS Appl. Electron. Mater*. **3**, 1127-1132 (2021).

[17] M. Naamneh, A. Prakash, E. B. Guedes, W. H. Brito, M. Shi, N. C. Plumb, B. Jalan, and M. Radović, “Surface state at BaSnO$_3$ evidenced by angle-resolved photoemission spectroscopy and *ab initio* calculations,” *arXiv*:2101.03399, 2021.

2020 publications

[18] V. R. S. K. Chaganti, T. K. Truttmann, F. Liu, B. Jalan, and S. J. Koester, “SrSnO$_3$ field-effect transistors with recessed gate electrodes,” *IEEE Elect. Dev. Lett*. **41**, 1428-1431 (2020).

2019 publications

[19] T. Truttmann, A. Prakash, J. Yue, T. E. Mates, and B. Jalan, “Dopant solubility, and charge compensation in La-doped SrSnO$_3$ films,” *Appl. Phys. Lett*. **115**, 152103 (2019).

[20] J. Yue, L. R. Thoutam A. Prakash, T. Wang, and B. Jalan, “Unraveling the effect of electron electron interaction on electronic transport in high-mobility stannate films,” *Appl. Phys. Lett.* **115**, 082102 (2019)

[21] U. S. Alaan, F. J. Wong, J. J. Ditto, A. W. Robertson, E. Lindgren, A. Prakash, G. Haugstad, P. Shafer, A. T. N’Diaye, E. Arenholz, B. Jalan, N. D. Browning, and Y. Suzuki, “Magnetism and transport in transparent high-mobility BaSnO$_3$ films doped with La, Pr, Nd and Gd,” *Phys. Rev. Mater*. **3**, 124402 (2019).

[22] A. Prakash, N. F. Quackenbush, H. Yun, J. Held, T. Wang, T. Truttmann, J. M. Ablett, C. Weiland, T.-L. Lee, J. C. Woicik, K. A. Mkhoyan and B. Jalan, “Separating electrons and donors in BaSnO$_3$ via band engineering,” *Nano Lett*. **19**, 8920-8927 (2019).

[23] A. Prakash, and B. Jalan, “Wide bandgap perovskite oxides with high room-temperature electron mobility,” *Adv. Mater. Interfaces* **6**, 1900479 (2019).

Invited talks/Seminars/Colloquiums related to project

1. S. J. Koester, "Self-Heating in Stannate Perovskite Field Effect Transistors," Workshop on Compound Semiconductor Materials and Devices (WOCSEMMAD 2022), Destin, FL, Feb. 20-23, 2022.
2. B. Jalan, "Band-engineered Alkaline-Earth Stannate Heterostructures," Oxide-based Materials and Devices International Conference XIII (Conference OE108), 2022 SPIE-Photonics West, San Francisco, CA, Jan. 22-27, 2022.
3. B. Jalan, "Navigating Novel Approaches to Atomically Precise Synthesis of "Stubborn" Metal Oxides," International AVS Symposium, Charlotte, NC, Oct. 25-28, 2021.
4. B. Jalan, "Self-Assembled Periodic Nanostructures Using Martensitic Phase Transformations," Materials Science &Technology Meeting, Columbus, OH, Oct. 18-22, 2021.
5. B. Jalan, "Navigating Novel Approaches to Atomically Precise Synthesis of "Stubborn" Metal Oxides," Department Seminar, Department of Materials Science and Engineering, Cornell University, Ithaca, NY, Sep. 30, 2021.
6. B. Jalan, "Modulation Doping in Alkaline Earth Stannates," 2021 DPG Meeting (virtual), Technical University of Dresden, September, 2021.
7. B. Jalan, "Remote Epitaxy and Strain Relaxation in $SrTiO_3$ using Hybrid MBE," Epitaxy on 2D materials for Layer Release and Their Applications, MIT, Boston, MA, Jun. 28-30, 2021.
8. B. Jalan, "Hybrid MBE growth of 'stubborn' metals and metal oxides," Workshop on MBE and Hybrid MBE Studies, Advanced Photon Source, Chicago, USA, May, 2021.
9. B. Jalan, "Innovation in synthesis approaches for atomically precise materials," Virtual department seminar, Max-Planck Institute of Microstructure Physics, Halle, Germany, May 6, 2021.
10. B. Jalan, "Wide bandgap perovskite oxides for power electronics," Department Seminar (virtual), EE Department, IIT Kanpur, India October 2020.
11. B. Jalan, "Wide bandgap perovskite oxides for power electronics," Department Colloquium (virtual), Physics Department, CUSB, India, October 2020.
12. S. J. Koester, "Ultra-wide-gap stannate perovskite transistors and process technology," Workshop on Compound Semiconductor Materials and Devices (WOCSEMMAD 2020), Palm Springs, CA, Feb. 16-19, 2020
13. B. Jalan, "Stabilization of High-temperature Polymorphs of $SrSnO_3$ at Room Temperature via Epitaxy" TMS Annual Meeting, San Diego, February, 2020.
14. B. Jalan, "Modulation Doping in Alkaline Earth Stannates" Electronic Materials and Applications (EMA), Orlando, Florida, January, 2020.
15. B. Jalan, "Wide bandgap perovskite oxides for power electronics" XXth International Workshop on Physics of Semiconductor Devices: IWPSD, Kolkata, India, December, 2019.
16. B. Jalan, "Where do defects in materials come from and whether we embrace them" Plenary Talk at the Atomically-Precise Materials Workshop, PNNL, Richland, WA, November, 2019.
17. B. Jalan, "Lead-Free Ferroelectric Thin Films for Waste Heat to Energy Conversion" 19th US-Japan Seminar on Dielectrics and Piezoelectric Ceramics, National Institute Advanced Industrial Science and Technology (AIST), Tsukuba, Japan, November, 2019.

18. B. Jalan, “Wide bandgap perovskite oxides for power electronics” ECE Colloquium, Department of Electrical and Computer Engineering, University of Minnesota, Minneapolis, October, 2019.
19. B. Jalan, “Where do defects in materials come from and do we embrace them?” Physics Colloquium, School of Physics and Astronomy, University of Minnesota, Minneapolis, October, 2019.
20. B. Jalan, “Radical MBE Approach for the growth of Metal Oxides of Stubborn Metals” Physical Electronics Symposium, Minneapolis, September, 2019.
21. B. Jalan, “Where do defects in materials come from and do we embrace them?” MN AVS Symposium, Minneapolis Minnesota, September, 2019.
22. B. Jalan, “Radical-based MBE Approach for Stubborn Metal Oxides” 19th International Conference on Crystal Growth and Epitaxy (ICCGE), Keystone, CO, July, 2019.

Honors / Awards

Koester elevated to Fellow of Optica (formerly OSA), 2021.

Koester named Russell J. Penrose Professor in Nanotechnology, 2021.

Jalan awarded Peter Mark Memorial Award from the American Vacuum Society, 2021.

Koester named Director of Minnesota Nano Center, 2021.

Jalan awarded TechConnect Innovation Award, 2020.

Jalan named Presidential Early Career Awards for Scientists and Engineers (PECASE), 2019.

Koester named Louis John Schnell Professor in Electrical and Computer Engineering, 2019.

List of Significant Collaborators related to project

Domestic Collaborators

Berardi Sensale Rodriguez, U of Utah - THz measurements and plasmonic devices; Yuri Suzuki, Stanford University - Magnetism; Chang-Beom Eom, U W Madison – Oxide heterostructures; Scott Chambers and Peter Sushko, PNNL – X-ray photoemission spectroscopy and DFT; Joel Ager, UC Berkeley – Seebeck measurements and electronic transport modeling; Yohannes Abates, University of Georgia – Nanoscale optical imaging; Richards James, University of Minnesota – Theory of phase transformation; Dr. Shin Mou and Adam Neal, AFRL – high temperature transport and capacitance spectroscopy; Jeehwan Kim, MIT, USA – Integration of oxides with 2D semiconductors; Andrea Alu, CUNY, NY – nanophotonics; Marco Bernardi, Caltech – first principle calculations of transport; David Gold-Haber Gordon – Stanford University – nano scale mK transport), Uttam Singisetti, University of Buffalo – DFT calculation of electronic transport; Sriram Krishnamoorty, UCSB – MOCVD growth of β-Ga_2O_3 films; Sang-Hyun Oh, University of Minnesota – Optical characterization of oxide semiconductors, Uttam Singisetti, University of Buffalo – High-speed pulsed measurements of stannate structures to evaluate self-heating and velocity saturation, Anirudha Sumant, Argonne National Laboratory – Deposition of nano-crystalline diamond on $SrSnO_3$ thin films in order to mitigate self-heating effects, Andrei Osinksy and Fikadu Alema, Agnitron Technology – Investigation of β-Ga_2O_3 heterostructure devices, Georges Pavlidis, University of Connecticut – Scanning thermal imaging of $SrSnO_3$ devices

International Collaborators

Roger De Souza, RWTH Aachen University, Germany – Ionic transport in stannate films; George Sawatzky, University of British Columbia, Canada – MBE growth and Photoemission; Kookrin Char, Seoul National University, Korea – Hybrid MBE growth of modulation doped structures; Milan Radovic, Paul Scherrer Institute (PSI), Switzerland – ARPES measurements of stannates; Hanus SEINER, the Institute of Thermomechanics, Czech Republic – Elastic property measurements of oxides; Juan Maria Garcia Lastra, Denmark Technical University, Denmark – DFT calculations; David Keeble, University of Dundee, UK – Positron Annihilation study of point defects; Eckhard Quant, University of Kiel, Germany – Diffraction and phase transformation; Prof. Subhananda Chakrabarti, IIT Bombay, India – PL measurements of defect states in stannates films

Students supported

Tristan Truttmann (Jalan), Fengdeng Liu (Jalan), Sreejith Nair (Jalan), Jin Yue (Jalan), Prakash Palamedu Sundaram (Koester), Prafful Golani (Koester), Jiaxuan Wen (Koester), Chin-Hsiang Liao (Koester), Saran Kumar Chaganti (Koester)