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Ultra High-Density Complex Oxide Quantum Wells for Emerging THz Plasmonic Effects and Devices

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14. ABSTRACT
This project aimed to develop novel complex oxide quantum wells with high electron densities and high mobility for fundamental study and for potential THz device applications. We proposed to exploit these structures by conducting detailed structural and electronic transport studies in order to understand, and eventually control, specific defects, local structure, and electronic transport in oxide heterostructures. Structures incorporating two-dimensional electron
gases (2DEGs) were emphasized. Education and research opportunities for graduate and undergraduate students were also emphasized. Stannate perovskites became the central focus. This project resulted in 20 peer-reviewed publications and two US patent disclosures.

15. SUBJECT TERMS
broken type-II band alignment, quantum well, heterostructure, THz, BaSnO3

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Ultra High-Density Complex Oxide Quantum Wells for Emerging THz Plasmonic Effects and Devices

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06/01/2016 – 05/31/2019

As outlined in the proposal, the overall goal of this project is to develop novel complex oxide quantum wells with high electron densities and high mobility for fundamental study and for potential THz device applications. We proposed to exploit these structures by conducting detailed structural and electronic transport studies in order to understand, and eventually control, specific defects, local structure, and electronic transport in oxide heterostructures. Structures incorporating two-dimensional electron gases (2DEGs) were emphasized. Education and research opportunities for graduate, and undergraduate students were also emphasized.

A Brief Summary of Accomplishments

Progress during the award period has been substantial. This award resulted in *20 peer-reviewed publications*, *2 US patents*, about two-dozen invited talks/colloquiums and a several prestigious awards. Progress has been accomplished in the four major areas:

Innovation in Synthesis Science, Novel Materials Development, and Advanced Characterizations

1. Perfection of the hybrid molecular beam epitaxy (MBE) growth of very precisely stoichiometric thin films and heterostructures of $NdTiO_3/SrTiO_3$ with tunable electron density [1]
2. Perfection of the growth of highly perfect wide band gap semiconductor, $BaSnO_3$ films using novel radical-based hybrid MBE with potential for realizing high room temperature electron mobility [2]
3. Exploration of band-offsets, and chemical doping control of $BaSnO_3$ films, a logical step on the path to band engineered quantum well structures [3,4]
4. First measurement of Dielectric measurements of $BaSnO_3$ films towards functional devices [5]
5. Hybrid MBE growth of a new wide bandgap $SrSnO_3$ films with record high electron mobility at room temperature [6]
6. *First* demonstration of a novel strain-stabilized phases of $SrSnO_3$ films [7]
7. Detailed study of transport properties using chemical dopants, and electrostatic doping of $SrSnO_3$ films [8,9]
8. *First* demonstration of modulation doping approach in a non-STO perovskite structure [10]
9. Combined experimental and computational study of the microstructure of $BaSnO_3$ films [11,12]

Emerging Phenomena and Discovery

10. Demonstration of high Terahertz (THz) conductivity in ultra-high density 2DEGs in $NdTiO_3/SrTiO_3$ heterostructures, an essential pre-requisite to develop high frequency plasmonic devices [13]
11. Discovery of novel ferromagnetism and giant magneto-resistance in MBE-grown defect-managed $NdTiO_3/SrTiO_3$ heterostructures [14]
12. Discovery of electron-electron interaction effects in La-doped $SrSnO_3$ films [15]

Devices Demonstration

13. THz characterization, and demonstration of first visible-transparent/terahertz-functional electromagnetic structures in ultra-conductive La-doped $BaSnO_3$ films [16]
14. Demonstration of the first depletion-mode MOSFET using all-epitaxial $BaSnO_3/SrTiO_3$ heterostructures [17]
15. *First* demonstration of metal-semiconductor FET (MESFET) using doped-$SrSnO_3$ [18]

Review Articles, Book Chapter and Commercialization

16. Patents, Invited review article and a book chapter on the development of wide bandgap perovskite [19-22]

List of Publications

[1] P. Xu, Y. Ayino, C. Cheng, V. Pribiag, R. Comes, P. V. Sushko, S. Chambers, and B. Jalan "Predictive control over charge density in the two-dimensional electron gas at the polar/non-polar $NdTiO_3/SrTiO_3$ interface", *Phys. Rev. Lett.* **117**, 106803 (2016).

[2] A. Prakash, P. Xu, X. Wu, G. Haugstad, X. Wang, and B. Jalan "Adsorption-controlled growth and the influence of stoichiometry on electronic transport of hybrid molecular beam epitaxy-grown $BaSnO_3$ films" *J. Mater. Chem. C* **5***,* 5730 (2017) (*Invited article*)

[3] S. A. Chambers, T. C. Kaspar, A. Prakash, G. Haugstad, B. Jalan, "Band alignment at epitaxial $BaSnO_3/SrTiO_3(001)$ and $BaSnO_3/LaAlO_3(001)$ heterojunctions" *Appl. Phys. Lett.* **108**, 152104 (2016).

[4] A. Prakash, P. Xu, A. Faghaninia, S. Shukla, J. W. Ager, C. S. Lo, and B. Jalan "Wide Band-gap Oxide with Room Temperature Conductivity Exceeding 10^4 S/cm", *Nat. Commun*. **8**, 15167 (2017)

[5] W. Nunn, A. Prakash, A. Bhowmik, R. Haislmaier, J. Yue, J. M. Garcia Lastra, and B. Jalan, "Frequency- and temperature-dependent dielectric response in hybrid molecular beam epitaxy-grown $BaSnO_3$ films" *APL Mater.* **6**, 066107 (2018)

[6] T. Wang, L. R. Thoutam, A. Prakash, G. Haugstad, and B. Jalan "Defect-driven Localization Crossovers in MBE-Grown La-doped $SrSnO_3$ films", *Phys. Rev. Mater.* (*Rapid Communication)* **1**, 061601(R) (2017)

[7] T. Wang, A. Prakash, Y. Dong, T. Truttmann, A. Bucsek, R. James, D. D. Fong, J.-W. Kim, P. J. Ryan, H. Zhou, T. Birol, and B. Jalan, "Engineering $SrSnO_3$ Phases and Electron Mobility at Room Temperature using Epitaxial Strain" ACS Appl. Mater. & Interfaces **10**, 43802 (2018)

[8] L. R. Thoutam, J. Yue, A. Prakash, T. Wang, K. Elangovan and B. Jalan,"Electrostatic control of insulator-metal transition in La-doped $SrSnO_3$ films" ACS. Appl. Mater. & Interfaces **11**, 7666 (2019)

[9] T. Truttmann, A. Prakash, J. Yue, T. E. Mates, and B. Jalan, "Dopant solubility, and charge compensation in La-doped $SrSnO_3$ films" **115**, 152103 (2019)

[10] A. Prakash, N. F. Quackenbush, H. Yun, J. Held, T. Wang, T. Truttmann, J. M. Ablett, C. Weiland, T-L. Lee, J. C. Woicik, KA. Mkhoyan and B. Jalan, "Separating electrons and donors in BaSnO3 via band engineering" Nano Lett. **19**, 8920 (2019)

[11] H. Yun, K. Ganguly, W. Postiglione, B. Jalan, C. Leighton, K. A. Mkhoyan, and J. S. Jeong, "Microstructure characterization of $BaSnO_3$ thin films on $LaAlO_3$ and $PrScO_3$ substrates from transmission electron microscopy" Sci. Rep. **8**, 10245 (2018)

[12] H. Yun, A. Prakash, B. Jalan, J. S. Jeong, KA Mkhoyan, "STEM beam channeling in BaSnO3/ LaAlO3 perovskite bilayers and visualization of 2D misfit dislocation network" Ultramicroscopy, **208**, 112863 (2019)

[13] S. Arezoomandan, A. Chanana, H. Condori, P. Xu, A. Nahata, B. Jalan, and B. S. Rodriguez, "Terahertz conductivity of ultra-high electron concentration 2DEGs in $NdTiO_3/SrTiO_3$ heterostructures" *APL Mater.* **4**, 076107 (2016).

[14] Y. Ayino, P. Xu, J. T.-Lazo, J. Yue, B. Jalan, and V. S. Pribiag "Spin-dependent transport at a ferromagnetic complex oxide interface", *Phys. Rev. Mater.* (*Rapid Communication)* **2**, 031401(R) (2018)

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

[16] S. Arezoomandan, A. Prakash, A. Chanana, J. Yue, A. Mao, S. Blair, A. Nahata, B. Jalan, and B. Sensale-Rodriguez "THz characterization and demonstration of visible-transparent/terahertz-functional electromagnetic structures in ultra-conductive La-doped $BaSnO_3$ Films", *Sci. Rep.* **8**, 3577 (2018)

[17] J. Yue, A. Prakash, M. C. Robbins, S. J. Koester, and B. Jalan, "Depletion Mode MOSFET using La-doped $BaSnO_3$ as a Channel Material" *ACS Appl. Mater. and Interfaces* **10**, 21061 (2018)

[18] V. R. S. K. Chaganti, A. Prakash, J. Yue, B. Jalan, and S. J. Koester, "Demonstration of a depletion-mode $SrSnO_3$ n-channel MESFET" *IEEE Elec. Dev. Lett.* **39**, 1381 (2018)

[19] A. Prakash and B. Jalan, Molecular beam epitaxy for oxide electronics. In: Molecular Beam Epitaxy: Materials and Applications for Electronics and Optoelectronics (ed. Hajime Asahi and Yoshiji Horikoshi), *John Wiley & Sons,* 423-452 (2019) (DOI: 10.1002/9781119354987.ch26)

[20] A. Prakash, and B. Jalan, "Wide Bandgap Perovskite Oxides with High Room-Temperature Electron Mobility" Adv. Mater. Interfaces **6**, 1900479 (2019) (Invited article)

Patents

[21] Richards James, and Bharat Jalan "The direct conversion of heat to electricity using phase transformations in ferroelectric oxides" US Patent App. 15/996,196 (2019)

[22] Abhinav Prakash, Tianqi Wang, and Bharat Jalan, "Highly Conducting Transparent Films" US Patent App. 16/125,187 (2019)

Invited talks/Seminars/Colloquiums

2016-2017

1. "Radical-based Oxide MBE and Electronic Transport of La-doped $BaSnO_3$ Thin Films" European MRS, Warsaw, Poland (September 2016)
2. "Transport in Band Engineered Complex Oxide Heterostructures" Physics Colloquium, Department of Physics, Auburn University (April 2017)
3. "Band Engineered Complex Oxide Interfaces" Compound Semiconductor Week (CSW), Berlin, Germany (May 2016)
4. "Hybrid MBE Growth of Highly Perfect Complex Oxide Thin Films and Heterostructures" Oxide Epitaxy Symposium, AACGE-21 Conference, Santa Fe, NM (August 2017)
5. "Two-Dimensional Electron Gas and Emerging Magnetism at Complex Oxide Interfaces", Department Seminar, Technical University of Denmark, Denmark (September 2017)

2017-2018

6. "MBE Growth, Structure, Defects and Transport in High-Mobility Stannate Films" Department Colloquium, Materials Department, UC Santa Barbara, Santa Barbara, CA, May 2018
7. "Novel Transparent Conducting Oxide with Room-Temperature Conductivity exceeding 10^4 S/cm" Oxide-based Materials and Devices IX, SPIE Photonics, San Francisco, CA, February 2018
8. "Novel Radical-based Hybrid Molecular Beam Epitaxy for Stannate Films and Heterostructures" Lawrence Symposium on Epitaxy, Scottsdale, AZ, February 2018
9. "Novel Radical-based Molecular Beam Epitaxy Approach for Metal Oxide Films Containing Elements of Low Oxidation Potential" Department Seminar, Department of Materials Science and Engineering, Penn State University, State College, PA, USA, October, 2017
10. "Atomically-Precise Layer Controlled Synthesis of Functional Oxides of Stubborn Metals" HESTEC Engineering and Science Symposium, Edinburg, Texas, October, 2017

11. "Novel Molecular Beam Epitaxy Approaxh for High Quality Perovskite Thin Films" Department Colloquium, Institute for Materials Science, Christian-Albrechts-Universitaet zu Kiel, Kiel, Germany, September, 2017
12. "Charge Transfer at Complex Oxide Interfaces" Department Seminar, Technical University of Denmark, Copenhagen, Denmark, September, 2017
13. "Novel Radical-based Molecular Beam Epitaxy Approach for Metal Oxide Films Containing Elements of Low Oxidation Potential" XXVI International Materials Research Congress, Cancun, Mexico, August, 2017
14. "Band-Engineered Complex Oxide Interfaces: Role of Defects and Growth Approaches" *Plenary talk*, 21st American Conference on Crystal Growth and Epitaxy, Santa Fe, NM, July, 2017
15. "Novel Radical-based Molecular Beam Epitaxy Approach for Metal Oxide Films Containing Elements of Low Oxidation Potential" 21st American Conference on Crystal Growth and Epitaxy, Santa Fe, NM, July, 2017

2018-2019

16. B. Jalan "Novel MBE Approaches for Atomically-Precise Synthesis of Metal Oxides for Stubborn Metals" International MBE Meeting, Shanghai, China September, 2018
17. B. Jalan "Electrostatic Control of Transport in Stannate Thin Films, Department Seminar, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Zhejiang, China, August, 2018
18. B. Jalan "Towards development of Oxide Heterostructures with high Room Temperature Mobility" Condensed Matter Physics Seminar, Department of Applied Physics, Stanford, CA, September 2018
19. B. Jalan "Making Oxidation Easier in Ultra High Vacuum" Materials Research Lecture, Department of Applied Physics and Materials Science, Caltech, CA, October 2018
20. B. Jalan "Making Oxidation Easier in Ultra High Vacuum" Department Seminar, Department of Materials Science and Engineering, MIT, Boston, October 2018
21. B. Jalan "Wide bandgap perovskite oxides for power electronics" Invited Colloquium, at HRL, Malibu, CA, October 2018
22. B. Jalan "Progress in Perovskite Oxide Films with High Room- Temperature Mobility" Invited talk, Physics and Chemistry of Surfaces and Interfaces (PCSI-46), Santa Fe, January, 2019
23. B. Jalan "MBE Growth and Electrostatic Control of Transport in Stannate Thin Films" Invited talk, Electronic Materials and Applications (EMA), Orlando, January, 2019
24. B. Jalan "Radical-based MBE Approach for Stannate Thin Films" Invited talk, 19th International Conference on Crystal Growth and Epitaxy (ICCGE), Keystone, CO, July, 2019

Awards

2019 Presidential Early Career Awards for Scientists and Engineers (PECASE)
2018 TechConnect Innovation award, Anaheim, California, USA
2017 AVS Paul Holloway Young Investigator Award, Tampa, Florida, USA
2017 American Association for Crystal Growth Young Author Award, Santa Fe, USA
2017 Emerging Young Investigator recognized by the Royal Society of Chemistry
2016 International MBE Young Investigator Award, Montpellier, France

List of Significant Collaborators

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; Yohannes Abate, UGA – Scanning probe characterizations.

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.