DOE/ER/45722-1

Final Report**For****Atomic Scale Chemical and Structural Characterization of
Ceramic Oxide Heterostructure Interfaces**

Submitted to:

Division of Materials Science and Engineering
Office of Basic Energy Sciences
Office of Science
U.S. Department of Energy

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5.29.03
Date

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1. Project Progress Summary

According to the original proposal, the research plan was divided into three tasks: (a) growth of oxide heterostructures for interface engineering using standard thin film deposition techniques, (b) atomic level characterization of oxide heterostructures using such techniques as STEM-Z combined with AFM/STM and conventional high-resolution microscopy (HRTEM) and (c) property measurements of aspects important to oxide heterostructures using standard characterization methods, including dielectric properties and dynamic cathodoluminescence measurements.

Each of these topics were further classified on the basis of type of oxide heterostructure. Type I oxide heterostructures consisted of active dielectric layers, including the materials $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ (BST), Y_2O_3 and ZrO_2 . Type II heterostructures consisted of ferroelectric active layers such as lanthanum manganate and Type III heterostructures consist of phosphor oxide active layers such as Eu-doped Y_2O_3 .

Accomplishments in Type I oxides:

- Deposition by *pulsed laser ablation* and optimization of various high and medium-K (dielectric constant) oxide films such as BST, Y_2O_3 , and ZrO_2 on Si substrates.
- Characterization of microstructure by atomic scale techniques, including HRTEM, XPS and XRR.
- Characterization and optimization of electrical properties (Capacitance-Voltage and Current Voltage).
- Elimination of thermodynamically stable interfacial SiO_2 layer by high temperature hydrogen ambient anneals to passivate dangling silicon bonds.
- Suppression of interfacial layer formation using silicon surface pre-nitridation prior to deposition of the films
- Nanometer scale chemical characterization of the interface for interfacial layer composition
- Close interaction with Motorola, Inc. and North Carolina A&T University for characterization of electrical properties.

Accomplishments in Type II oxides:

- Deposition by *pulse laser ablation* and optimization of various colossal magnetoresistive films including doped perovskite LMO ($\text{La}_{0.7}\text{MnO}_{3-\delta}$ doped with Ca, Ba, Sr, Pb) to act as a ferromagnetic biasing source to PBCMO ($\text{Pr}_{0.65}\text{Ba}_{0.05}\text{Ca}_{0.3}\text{MnO}_{3-\delta}$) on LaAlO_3 substrates.
- Characterization of atomic-scale microstructure by HRTEM, XRD and conventional TEM.
- Characterization of magnetic properties of the oxide films by SQUID
- Deposition and characterization of TMR (Tunneling Magnetoresistance) materials by *pulsed laser ablation* and *sputtering*.
- Enhanced MR ratios and lower transition temperatures of these oxides due to doping refinements in the microstructure.

Accomplishments in Type III oxides:

- Deposition by *pulsed laser ablation* and optimization of europium activated yttrium oxide ($\text{Eu:Y}_2\text{O}_3$) on various substrates, including quartz, sapphire, Si, and LaAlO_3 .
- Deposition of diamond buffer layers to enhance surface roughness of the films and consequently the optical properties.
- Characterization of interfaces using STEM-Z microscopy, HRTEM, XRD and XPS.
- Optical property characterization using cathodoluminescence (CL) and photoluminescence (PL).
- Development of a model that predicts the behavior of phosphor thin films based on numerous variables such as substrate type, film thickness, crystallinity, etc.
- Strong interactions with Motorola, Inc. and Oak Ridge National Laboratory in characterization and property measurement.

2. Relevant Papers

Several papers attributable to the work performed and published during the funded period are listed below.

- 1) K-G Cho, D. Kumar, Z. Chen, P. H. Holloway and R. K. Singh, "Modeling of Cathodoluminescent and Photoluminescent Properties of Pulsed Laser Deposited Eu-doped Y₂O₃ Films," *MRS Symposium Proceedings*, **558**, 21-27 (1999).
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- 5) S. Pietambaram, D. Kumar, R. K. Singh, C. B. Lee and V. S. Kaushik, "Microstructure, Magnetoresistance and Magnetic Properties of Pulsed-laser-deposited External, Internal and Mixed-doped Lanthanum Manganite Films," *Journal of Applied Physics*, **86**, 6, 3317-3326 (1999).
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- 7) V. Craciun, A. Srivastava, J. Howard, R. K. Singh and J. Perriere, "Characteristics of Ba_{0.5}Sr_{0.5}TiO₃ Thin Films Grown by Ultraviolet-assisted Pulsed Laser Deposition," *Applied Physics Letters*, **A**, **69**, S787-789 (1999).
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- 10) A. Srivastava, V. Craciun, J. Howard and R. K. Singh, "Enhanced Electrical Properties of Ba_{0.5}Sr_{0.5}TiO₃ Thin Films Grown by Ultraviolet-assisted Pulsed Laser Deposition," *Applied Physics Letters*, **75**, 3002-3004 (1999).
- 11) V. Craciun, J. Howard and R. K. Singh, "Ultraviolet-assisted Pulsed Laser Deposition of Thin Oxide Films," *MRS Proceedings*, **S74**, 193-198 (1999).
- 12) D. Kumar, K-G Cho, Z. Chen, V. Craciun, P. H. Holloway and R. K. Singh, "Electroluminescent Characteristics of Pulsed Laser Deposited Epitaxial Eu-doped Y₂O₃ Thin Films," *MRS Proceedings*, **S74**, 11-17 (1999).
- 13) V. Craciun, J. Perriere, J. Howard, D. Craciun and R. K. Singh, "Characteristics of ZnO Films Grown by Ultraviolet-assisted Pulsed Laser Deposition," *MRS Fall Meeting*, Boston (1999).
- 14) V. Craciun, J. Howard, A. Srivastava, R. K. Singh and J. Perriere, "Low Temperature Growth of Barium Strontium Titanate Films by Ultraviolet-assisted Pulsed Laser Deposition," *MRS Fall Meeting*, Boston (1999).
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- 21) V. Craciun, J. Howard and R. K. Singh, "Enhanced Properties of UV Assisted Deposition of High K Dielectrics," *Proceeding of the 10 Workshop on Physics of Solid State Devices*, Delhi, (2000).
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- 28) A. Srivastava, D. Kumar, R. K. Singh, H. Venkataraman and W. R. Eisenstadt, "Improvement in Electrical and Dielectric Behavior of $(\text{Ba,Sr})\text{TiO}_3$ Thin Films by Ag Doping," *Physical Review*, **B61** (11) 7305-7307 (2000).
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- 31) S. Pietambaram, D. Kumar, R. K. Singh and C. B. Lee, "Magnetotransport and Magnetic Properties of $\text{La}_{0.7}\text{MnO}_{3-\delta}$ and $\text{Pr}_{0.55}\text{Ba}_{0.05}\text{Ca}_{0.3}\text{MnO}_{3-\delta}$ Superlattices," *Applied Physics Letters*, **78**, 243-245 (2001).
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- 33) D. Singh, V. Craciun, R. Houriet, R. K. Singh, R. Vacassy and H. Hofmann, "Influence of Thin Film Microstructure, Particle Morphology and Crystallinity of Electrochemical Kinetics of LiMnO_4 Electrodes for Li Ion Rechargeable Batteries," *Journal Power Sources*, **97-8**, 826-831 (2001).
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3. Students Supported Under this Grant

The students currently supported by this grant are:

Nabil Bassim – University of Florida – Ph.D. – 12/2002 grad. date
Joshua Howard – University of Florida – Ph.D. – 12/2002 grad. Date
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4. Post Doctoral Researchers supported under this grant

D. Kumar – University of Florida
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