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The 24th annual meeting will address interdisciplinary topics including neuromorphic computation; ionic and electrochemical modification of electronic materials; nonequilibrium states and optical control; and oxide integration into energy and logic platforms.

Accomplishments: Conference was a success.

Training Opportunities: Nothing to Report

Results Dissemination: Nothing to Report

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report



iWOE 24 - 2017
International Workshop on Oxide Electronics
Sunday, September 24 - Wednesday, September 27
Chicago, Illinois, USA

The International Workshop on Oxide Electronics (iWOE) is the premier meeting focused on recent advances and emerging trends in the field of multifunctional complex oxides and their utilization in electronic devices.

The 24th annual meeting will address interdisciplinary topics including neuromorphic computation; ionic and electrochemical modification of electronic materials; nonequilibrium states and optical control; and oxide integration into energy and logic platforms.



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2017 iWOE Presentation Schedule

Monday, September 25

Session 1: Electronic Structure (chair: Lane Martin)

8:30-9:00 Chris Van de Walle (*Univ. California, Santa Barbara*)

Electronic and Optical Properties of Rare Earth Titanates

9:00-9:30 Scott Chambers (*Pacific Northwest National Lab.*)

High-Energy Resolution X-Ray Photoemission and Ultraviolet Photoemission as Probes of Electronic Structure at Complex Oxide Heterostructures

9:30-9:45 Gennadii Laskin (*Max Planck Institute for Solid State Research*)

SrRuO₃ as a Model System for Quantum Dots of Correlated Materials

9:45-10:00 Robert Green (*Univ. British Columbia*)

Resonant X-ray Reflectometry of Oxide Heterostructures

10:00-10:30 Kyle Shen (*Cornell Univ.*)

Controlling Electronic Structure through Epitaxial Strain in Oxide Thin Films

Session 2: Non-Equilibrium Behavior (chair: Divine Kumah)

10:45-11:15 Richard Averitt (*Univ. California, San Diego*)

Ultrafast Dynamics and Control in Transition Metal Oxides: Recent results on manganites and cuprates

11:15-11:45 Nicole Benedek (*Cornell Univ.*)

Shedding Light on Materials Out of Equilibrium: Ultrafast control of complex oxide properties from first principles

11:45-12:00 Jason Hoffman (*Harvard Univ.*)

Unconventional Slowing Down of Electronic and Structural Dynamics in Photoexcited Charge-Ordered La_{1/3}Sr_{2/3}FeO₃

12:00-1:30 Lunch

1:30-3:00 Poster Session 1

Session 3: New Routes to Functional Behavior (chair: Nicole Benedek)

3:15-3:45 Matthew Rosseinsky (*Univ. Liverpool*)

Crystal Chemistry and Computation in the Design and Discovery of Oxide Materials and Interfaces

3:45-4:00 Julia Mundy (*Univ. California, Berkeley*)

Functional Electronic Inversion Layers at Ferroelectric Domain Walls

4:00-4:15 Ho Nyung Lee (*Oak Ridge National Lab.*)

Neel Skyrmion Lattice Stabilized in Iridate-Manganite Heterostructures

4:15-4:45 Venkat Gopalan (*Penn State Univ.*)

Polar Metal Oxides: Science and applications

4:45-5:00 Josep Fontcuberta (*Institut de Ciència de Materials de Barcelona*)

Reversing ON/OFF Resistance States in BaTiO₃ Ferroelectric Tunnel Junctions: Entangled role of polarization and ionic motion

Session 4: Metal-Insulator Transitions (chair: Marta Gibert)5:15-5:45 Susanne Stemmer (*Univ. California, Santa Barbara*)

Metal-Insulator Transitions at Oxide Interfaces

5:45-6:00 Danfeng Li (*Stanford Univ.*)Metal-Insulator Transitions in Freestanding NdNiO₃ Films6:00-6:30 Philippe Ghosez (*Univ. Liège*)

On the Origin of the Metal-Insulator Transition in Rare-Earth Nickelates

Tuesday, September 26**Session 5: Defects, Ionics, and Functional Properties** (chair: Bharat Jalan)8:30-9:00 Sverre Selbach (*Norwegian Univ. of Science and Technology*)

Local Strain Fields and Point Defects in Epitaxial Thin Films of Ferroic Oxides

9:00-9:30 Alex Frano (*Univ. California, Berkeley*)

Disorder-Free Electrochemical Doping of Oxygen Charge Carriers into Epitaxial Metal-Oxide Films

9:30-9:45 Natalie Dawley (*Cornell Univ.*)Defect Mitigating (SrTiO₃)_n(BaTiO₃)_mSrO Superlattices for mmWave Tunable Dielectrics9:45-10:00 Mark Huijben (*Univ. Twente*)Enhanced Lithium Transport in Highly Ordered LiMn₂O₄ Cathode Films Towards Solid-State Batteries10:00-10:15 Jonathan Hwang (*MIT*)Interaction of CO₂ and NO_x Gas on Transition Metal Perovskites10:15-10:30 Martina Müller (*Research Center Jülich*)2D Electron System at the Magnetically Tunable EuO/SrTiO₃ interface**Session 6: Field-Effect Gating and Devices** (chair: Ambrose Seo)10:45-11:15 Chris Leighton (*Univ. Minnesota*)Control of Ferromagnetism in Ion-Gel-Gated La_{1-x}Sr_xCoO₃ Thin Films11:15-11:45 Suman Datta (*Notre Dame Univ.*)

A New Computing Substrate Enabled by Functional Oxides

11:45-12:00 Takeaki Yajima (*Univ. Tokyo*)Designing Type II Neurons via Instability of VO₂ Metal-Insulator Transition

12:00-1:30 Lunch

1:30-3:00 Poster Session 2

Session 7: Materials Design and Synthesis (chair: Gertjan Koster)3:15-3:45 T. Saha-Dasgupta (*S.N. Bose National Centre*)

3d-5d Double Perovskites: Playground for Oxide Electronics

3:45-4:15 Yuefeng Nie (*Nanjing Univ.*)

In Situ Observation of Layer-By-Layer Mean Inner Potential Oscillations and Precise Growth Control of Oxide Interfaces

- 4:15-4:30 David Harris (*Univ. Wisconsin*)
 Suppressed Disorder in Superconducting $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ Epitaxial Thin Films
- 4:30-5:00 Yuichi Shimakawa (*Kyoto Univ.*)
 Control of Oxygen Coordination Environment in Transition-Metal Oxides
- 5:00-5:15 Sohrab Ismail-Beigi (*Yale Univ.*)
 Unusual Orbitally-Polarized Insulating State at a Titanate/Cobaltate Interface
- 5:15-5:30 Makoto Minohara (*High Energy Accelerator Research Organization*)
 Growth of Antiperovskite-Type Oxide Ca_3SnO Thin Films by Pulsed Laser Deposition
- 6:30 Conference Banquet
Boarding begins at 6:30pm and ends promptly at 7:00pm

Wednesday, September 27

Session 8: Topology and Ferroics (chair: Steve May)

- 8:30-8:45 Padraic Shafer (*Lawrence Berkeley National Lab.*)
 In-Plane Spin Cycloid Structure in Multiferroic BiFeO_3 Thin Films
- 8:45-9:00 Sungmin Park (*Seoul National Univ.*)
 Selective Control of Multiple Ferroelectric Switching Pathways Using Trailing Flexoelectric Field
- 9:00-9:30 Masashi Kawasaki (*Univ. Tokyo*)
 Topological Functions in Oxide Heterostructures
- 9:30-9:45 Marta Gibert (*Univ. Geneva*)
 Unusual Magnetism in LaNiO_3 -Based Superlattices
- 9:45-10:00 Ingrid Hallsteinsen (*Lawrence Berkeley National Lab.*)
 Controlling the Magnetic Spin Reconstruction in (111)-Oriented $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{LaFeO}_3$ Heterostructures

Session 9: Interfacial Electronic Behavior (chair: Julia Mundy)

- 10:15-10:45 Guus Rijnders (*Univ. Twente*)
 Tuning the Properties of Oxide Heterostructures by Interfacial Oxygen Octahedral Coupling
- 10:45-11:15 Kookrin Char (*Seoul National University*)
 Manipulating $\text{LaInO}_3/\text{BaSnO}_3$ Polar Interface
- 11:15-11:30 Ryan Comes (*Auburn Univ.*)
 Surface and Interfacial Phenomena in the $\text{LaFeO}_3/\text{n-SrTiO}_3$ Heterojunction
- 11:30-11:45 Ke Zou (*Yale Univ.*)
 Revealing Topological States at a Hybrid Oxide Chalcogenide Interface
- 11:45-12:15 Marc Gabay (*Université Paris-Sud*)
 Multifunctional Electronic States of 2DEGs at Perovskite Oxide Surfaces
- 12:15-12:30 Awards and Closing

Electronic and Optical Properties of Rare Earth Titanates

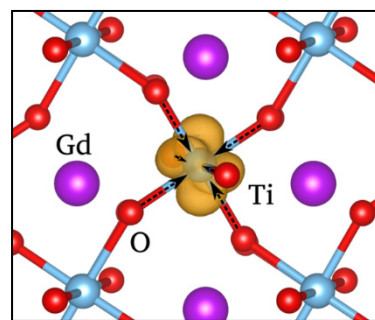
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The rare-earth titanates (RTiO_3 , where R is a rare-earth atom) have become the focus of great interest because of their use in complex-oxide heterostructures that display two-dimensional electron gases (2DEGs) with unprecedented high densities [1]. These compounds are Mott insulators, with a Mott-Hubbard gap opening up within the Ti 3d states. This gap is commonly reported to be 0.2-0.7 eV across the series. The values are based on optical reflectivity measurements, from which the onset of optical absorption is derived. We demonstrate that the Mott-Hubbard gap of GTO is actually much larger, close to 2 eV. This conclusion is based on both first-principles calculations (using hybrid density functional theory) and photoluminescence (PL) measurements, which display a strong peak near 1.8 eV and an onset in PL excitation at about the same energy [2].

The experimentally observed onset in absorption is not related to the gap, but is associated with small polarons. We find an excellent match between first-principles calculations of small-polaron related optical transitions and optical conductivity measurements on Sr-doped GTO [3], where the Sr doping introduces small hole polarons. We have also performed a study of other impurities and point defects that may affect the conductivity [4]. Among native defects, the cation vacancies have the lowest formation energies in oxygen-rich conditions, and oxygen vacancies have the lowest formation energy in oxygen-poor conditions. Among the impurities, Sr_{Gd} , H_i and C_o are easily incorporated. The defects and impurities are intrinsically stable only in a single "natural" charge state, to which various numbers of hole polarons can be bound, which explains the frequent observation of *p*-type hopping conductivity in the rare-earth titanates.



Atomic structure and spin density for a hole polaron in GdTiO_3 .

Given the similarities in electronic structure between the rare-earth titanates, our results for GTO have repercussions for the other members of the series. The results also affect the design of complex-oxide heterostructures involving these materials.

Work performed in collaboration with Lars Bjaalie, Anderson Janotti, Burak Himmetoglu, Karthik Krishnaswamy and supported by NSF, LEAST (one of six centers of STARnet, a Semiconductor Research Corporation program sponsored by MARCO and DARPA), and ONR.

References:

1. P. Moetakef *et al.*, Electrostatic carrier doping of $\text{GdTiO}_3/\text{SrTiO}_3$ interfaces, *Appl. Phys. Lett.* **99** (2011) 232116.
2. L. Bjaalie *et al.*, Determination of the Mott-Hubbard gap in GdTiO_3 , *Phys. Rev. B* **92** (2015) 085111.
3. L. Bjaalie *et al.*, Small hole polarons in rare-earth titanates, *Appl. Phys. Lett.* **106** (2015) 232103.
4. L. Bjaalie *et al.*, Point defects, impurities, and small hole polarons in GdTiO_3 , *Phys. Rev. B* **93** (2016) 115316.

High-energy resolution x-ray photoemission and ultraviolet photoemission as probes of electronic structure at complex oxide heterostructures

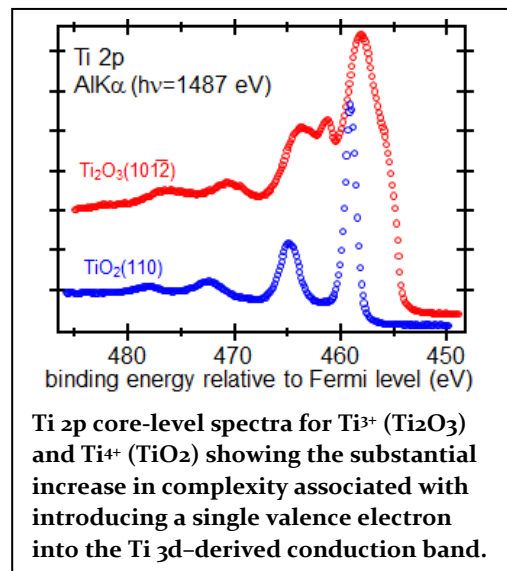
Scott A. Chambers

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Nucleation and epitaxial film growth during complex oxide heterostructure formation involves the simultaneous occurrence of several physical and chemical processes. These include metal and oxygen adatom arrival at and diffusion along the substrate surface, local chemistry between adatoms, as well as between adatoms, substrate atoms and defects, and condensation and crystallization of new atomic layers. Local electronic structures evolve into bands as heterostructures form. Throughout these complex processes, it is very useful to track the electronic evolution of specific elements within the different layers as well as the eventual formation of bands as layers are completed. Band alignment and the magnitudes and signs of built-in potentials within the individual layers are also of considerable interest as these properties directly affect electronic transport in completed heterostructures.

Core-level and valence band x-ray and ultraviolet photoelectron spectroscopy (XPS/UPS) are invaluable tools for these purposes, especially when measured with high energy resolution and *in situ*. Considerable insight can be gained into the detailed electronic and magnetic properties of transitional metal (TM) cations within the local structural environments that exist in heterostructures. The line shapes of core photoelectron peaks from TM cations in oxides are often quite complex relative to those for pure TM phases due to changes in valence, coupling of angular momenta between core holes and unpaired *d* electrons in the valence or conduction bands, and many-body effects resulting from charge transfer between O ligands the TM cations during core photoionization. However, advances in *ab initio* relativistic Hartree-Fock theory allow these line shapes to be calculated without adjustable parameters. Once these line shapes are understood, combining core and valence electron binding energies for constituent oxides and their heterostructures allows band alignment and built-in potentials to be accurately determined.



Ti 2p core-level spectra for Ti^{3+} (Ti_2O_3) and Ti^{4+} (TiO_2) showing the substantial increase in complexity associated with introducing a single valence electron into the Ti 3d-derived conduction band.

An interesting new development is the use of ultra-bright UV and hard x-ray light sources to probe bound electrons in defect states within the bandgaps of semiconducting oxides at the surface and in the sub-surface region. Much attention has been paid to the intense donor electron feature near the Fermi level in, for example, $n\text{-SrTiO}_3(001)$ that has been partially reduced via defect creation associated with interface formation or photon stimulated desorption of O. In contrast, relatively little effort has gone into understanding the weak, but ever-present photoemission intensity from the lower two thirds of the gap. Our tentative conclusion is that the intensity deep in the gap is a result of acceptors, possibly Sr vacancies, that form during tube furnace annealing.

In this talk, I will give an overview of these experimental and theoretical investigations, drawing from our experience using *in situ* XPS and UPS to characterize MBE-grown complex oxide heterostructures.

SrRuO₃ as a Model System for Quantum Dots of Correlated Materials

Gennadii Laskin^{1*}, Wolfgang Braun¹, Hans Boschker¹, and Jochen Mannhart¹

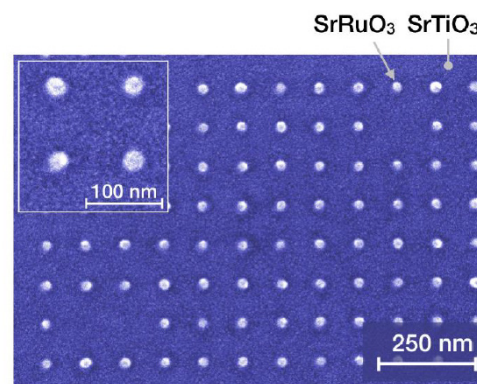
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For several decades quantum dots or artificial atoms have been attracting attention as zero-dimensional electron systems due to their remarkable electronic properties [1]. In conventional quantum dots made of semiconductors or metals, correlation effects, if existing, arise mainly from the quantum confinement induced for example by lateral patterns, superlattices, electric fields or magnetic impurities to Kondo-type behavior [2,3].

Here we propose and present artificial atoms fabricated from materials with inherently correlated electron systems [4]. Size-quantization effects generated by confinement of such materials into quantum dots may cause significant modifications of the already existing correlated electron system and lead to properties that are not present in other materials.

As a material for fabrication of artificial atoms we use SrRuO₃ [5] grown on SrTiO₃ (100). The quantum dots are grown by pulsed laser deposition and patterned by electron beam lithography with spatial resolution down to 20 nm.



An array of 20-nm-diameter SrRuO₃ dots imaged by SEM.

We will show that transforming the SrRuO₃ films into quantum dots enhances their magnetic properties.

References:

1. R. C. Ashoori, Electrons in artificial atoms, *Nature* **379** (1996) 413-419. DOI: 10.1038/379413a0
2. T. Chakraborty, Physics of the artificial atoms: quantum dots in a magnetic field, *Comments Cond. Mat. Phys.* **16** (1992) 35-68
3. M. Ternes, A. J. Heinrich, W.-D. Schneider, Spectroscopic manifestations of the Kondo effect on single adatoms, *J. Phys.: Condens. Matter* **21** (2008) 053001. DOI: 10.1088/0953-8984/21/5/053001
4. J. Mannhart, H. Boschker, T. Kopp, R. Valenti, Artificial atoms based on correlated materials, *Rep. Prog. Phys.* **79** (2016) 084508. DOI: 10.1088/0034-4885/79/8/084508
5. G. Koster, L. Klein, W. Siemons, G. Rijnders, J. S. Dodge, C.-B. Eom, D. H. A. Blank, M. R. Beasley, Structure, physical properties, and applications of SrRuO₃ thin films, *Rev. Mod. Phys.* **84** (2012) 253-298. DOI: 10.1103/RevModPhys.84.253

Resonant X-ray Reflectometry of Oxide Heterostructures

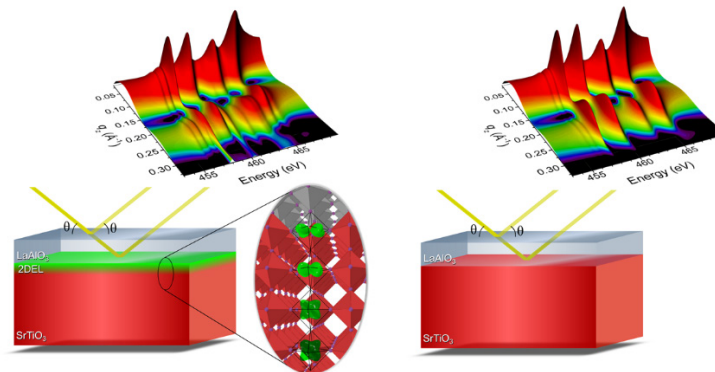
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Interfaces in oxide heterostructures exhibit a wide range of emergent phenomena, such as interface charge transfer, two dimensional electron gases (2DEGs), superconductivity, and ferromagnetism between non-magnetic materials [1]. Such phenomena result from the interface-induced tuning of the various spin, charge, orbital, and lattice degrees of freedom, and show great promise for electronics applications. However, while the emergent phenomena are readily apparent, obtaining electronic structure information specific to the nanometer-scale buried interface region—in order to understand and further tune the emergent phenomena—is an understandably difficult task.



Resonant x-ray reflectometry is strongly sensitive to the orbital symmetry and charge density profiles of an oxide 2D electron gas, as shown by the comparison between an interface with a 2DEG (left), and an interface with no 2DEG (right). A rich set of features are present in the spectrum on the left due to the interfacial reconstructions.

Here I will present our recent results of the study of oxide interface phenomena using resonant x-ray reflectometry, a new synchrotron-based spectroscopic technique that we have shown provides interface and depth-sensitive information on electronic and magnetic structure with spatial resolution at the level of atomic planes [2-4]. I will detail our recent results of extracting the high resolution depth profiles of charge density and orbital symmetry in a series of $\text{LaAlO}_3/\text{SrTiO}_3$ heterostructures. Further, I will show our results studying the interplay between electronic reconstruction and oxygen vacancies in trilayer systems such as $\text{LaAlO}_3/\text{LaMnO}_3/\text{SrTiO}_3$, where we have shown that exceptionally high electron mobilities can be realized through a unique modulation doping effect [4]. Resonant x-ray reflectometry is able to disentangle the presence of oxygen vacancies at the upper interface and pure electronic reconstruction at the lower interface, yielding crucial insight into the mechanism for high mobilities.

References:

1. Hwang *et al.*, *Nature Mater.* **11** (2012) 103. DOI: 10.1038/NMAT3223
2. Macke *et al.*, *Adv. Mater.* **26** (2014) 6554. DOI: 10.1002/adma.201402028
3. J. E. Hamann-Borrero *et al.*, *npj Quant. Mater.* **1** (2016) 16013. DOI: 10.1038/npjquantmats.2016.13
4. Z. Liao *et al.*, *Nature Mater.* **15**, 425 (2016). DOI: 10.1038/nmat4579
5. Y. Z. Chen *et al.*, *Nature Mater.* **14**, 801 (2015). DOI: 10.1038/nmat4303

Controlling Electronic Structure through Epitaxial Strain in Oxide Thin Films

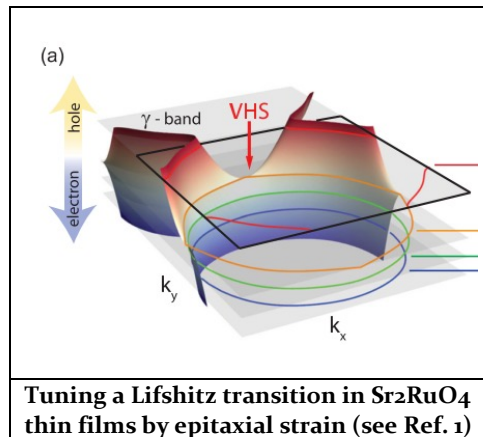
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Our ability to control the electronic structure of materials, for instance in semiconductors, has had enormous scientific and technological implications. Recently, this concept has been extended to complex oxides which can possess inherently strong quantum many-body interactions which can harbor novel electronic or magnetic properties. The ability to deterministically manipulate the electronic structure of complex oxides using tuning parameters such as epitaxial strain or dimensional confinement will be important to realizing future devices based on complex oxides. I will describe recent experiments using epitaxial strain to drive a Lifshitz transition in thin films of Sr_2RuO_4 and Ba_2RuO_4 , which also changes the sign of the low-temperature Hall coefficient by strain. I will also describe recent experiments using dimensional confinement in ultrathin complex oxide quantum wells to control ground states and mass enhancements.



The research reported here has been primarily supported by the Air Force Office of Scientific Research under FA9550-12-1-0335.

References:

1. B. Burganov, C. Adamo, A. Mulder, M. Uchida, P.D.C. King, J.W. Harter, D.E. Shai, A.S. Gibbs, A.P. Mackenzie, R. Uecker, M. Bruetzsch, M.R. Beasley, C.J. Fennie, D.G. Schlom, and K.M. Shen, Strain Control of Fermiology and Many-Body Interactions in Two-Dimensional Ruthenates. *Physical Review Letters* **116**, 197003 (2016).

Ultrafast dynamics and control in transition metal oxides: recent results on manganites and cuprates

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The macroscopic response of numerous correlated transition metal oxides is strongly influenced by nano-to-mesoscopic phenomena including phase coexistence that, in turn, arises from competing interactions. This manifests in the dynamics of these materials and can be probed – with the looming grand challenge of control – using ultrafast optical pulses. Two phenomena that have been extensively investigated using ultrafast optics include the insulator-to-metal transition (IMT) and superconductivity. In this talk, I will present an overview of our recent activities in these areas using time-resolved terahertz techniques on thin films and single crystals.

I will present results of our recent investigations on IMT dynamics of manganite ($\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$) thin films that have been strain-engineered to quench the thermal IMT. Strain yields, at all temperatures, a robust antiferromagnetic charge-ordered insulating phase resulting from orthorhombic distortion of the underlying octahedral framework. Short pulse photoexcitation initiates a nonthermal IMT to a persistent “hidden” ferromagnetic metallic phase. Our measurements reveal that the transition is cooperative and ultrafast, requiring a critical absorbed photon density to activate local charge excitations that mediate magnetic-lattice coupling stabilizing the metallic phase¹. Since the photoinduced changes are metastable, we have been able to image mesoscopic features of the photoinduced state using infrared nanoscopy correlated with simultaneous topographic and magnetic imaging using AFM and MFM. Shot-to-shot images indicate a boundary region with a decreased threshold for the photoinduced IMT².

In the cuprates, the c-axis electrodynamic response (i.e. the Josephson plasma resonance – JPR) of $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ ($x = 11.5\%$) has previously been investigated following near-infrared excitation, revealing spectroscopic hints of transiently enhanced superconductivity on a picosecond timescale at the expense of the striped order phase³. We have performed preliminary measurements under different excitation conditions (in comparison to Ref. [3]) that results in a novel long-lived (hundreds of picoseconds) plasma resonance that is dramatically blue-shifted from the static JPR. The nature and possible origins of this robust spectroscopic mode will be presented.

The work on $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ was supported by the Department of Energy Basic Energy Sciences (DOE-BES) under DE-SC0012375, while the work on $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ was supported by DOE-BES DE-SC0012592.

References:

1. J. Zhang, X. Tan, M. Liu, S. W. Teitelbaum, K. W. Post, F. Jin, K. A. Nelson, D. N. Basov, W. Wu, R. D. Averitt, Cooperative photoinduced metastable phase control in strained manganite films, *Nature Materials* **15**, 956 (2016).
2. A. S. McLeod, J. Zhang, et al., manuscript in preparation.
3. D. Nicoletti, E. Casandruc, Y. Laplace, V. Khanna, C. R. Hunt, S. Kaiser, S. S. Dhesi, G. D. Gu, J.P. Hill, A. Cavalleri, Optically induced superconductivity in striped $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ by polarization selective excitation in the near infrared, *Phys. Rev. B* **90**, 100503(R) (2014).
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Shedding light on materials out of equilibrium: Ultrafast control of complex oxide properties from first principles

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The recent development of intense ultrashort mid- and far-infrared laser sources has created an opportunity for functional materials based on the direct excitation of infrared active phonons. Strong excitation of infrared active phonons can produce sizable unidirectional distortions of crystal structure through non-linear couplings between various lattice modes. Complex oxides provide an important test-ground for this experimental approach due to their chemical diversity, strong coupling to optical fields, and demonstrated connection between subtle structural changes and functional properties. Early experiments in complex oxides are intriguing, suggesting that non-linear phonon coupling is responsible for transiently induced insulator-metal phase transitions and enhanced superconductivity in optical experiments.

In this talk, I will describe our recent theoretical efforts exploring selective control of functional properties in perovskite oxides that exploit non-linear lattice dynamics. Using first-principles techniques we show that optical control of various properties is experimentally feasible and that, when combined with epitaxial strain, it is possible to transiently stabilize and explore phases inaccessible in the equilibrium phase diagram.

This work was supported by the National Science Foundation under awards DMR-1550347 and DMR-1120296 (Cornell Center for Materials Research, an NSF MRSEC). This work made use of high-performance computing facilities provided by the Cornell Center for Advanced Computing and the National Science Foundation through XSEDE allocation DMR-160052.

Unconventional Slowing Down of Electronic and Structural Dynamics in Photoexcited Charge-Ordered $\text{La}_{1/3}\text{Sr}_{2/3}\text{FeO}_3$

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The intensive study of coupled phase transitions in strongly correlated materials has been motivated by interest in the fundamental physics and potential applications of emergent phenomena, such as high-temperature superconductivity, metal-to-insulator phase transitions, and colossal magnetoresistance. The complex interactions amongst the multiple degrees of freedom in correlated systems can be effectively disentangled on ultrafast time scales due to the distinct dynamics associated with different interaction mechanisms. Many studies of the transient response in correlated materials have therefore used optical, terahertz, x-ray, and electron probes to explore the initial response on sub-picosecond time scales following ultrafast excitation. On time scales beyond nanoseconds, it is often assumed that charge, spin, and lattice degrees of freedom have exchanged energy sufficiently to reach internal equilibrium, i.e., internal degrees of freedom of the system reach the same temperature. In this regime, the evolution of physical processes is primarily governed by the cooling of the system through thermal exchange with its external environment.

Here, we report on the non-thermal recovery of the charge ordered phase in photoexcited $\text{La}_{1/3}\text{Sr}_{2/3}\text{FeO}_3$ (LSFO) thin films. Using time-resolved optical spectroscopy and x-ray diffraction, we directly track three quantities in the time domain: i) the optical reflectivity, ii) the superlattice x-ray diffraction intensity as a result of charge ordering, and iii) the out-of-plane lattice constant of LSFO. When the sample temperature increases towards the charge-ordering temperature, we observe a concurrent increase in the recovery time of both the transient optical reflectivity and the charge-order diffraction peak intensity, which extends well-beyond the few-nanosecond thermal recovery of the lattice constant. We combine our experimental study with first principle DFT+U calculations to elucidate a microscopic picture of magnetic-interaction driven slowing down and suggest a pseudo-critical phenomenon close to a weakly first-order phase transition.

This research has been supported in part by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES) under Contracts No. DE-SC0012375 and No. DE-AC02-06CH11357.

Crystal chemistry and computation in the design and discovery of oxide materials and interfaces

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I will discuss recent progress in accelerating the discovery of materials by use of crystal chemically-informed computational tools (1,2) to focus the experimental search of compositional space. The application of crystal chemical considerations to the design of coherent interfaces between materials with different crystal structures to permit layer-by-layer heterostructure growth is also discussed (3). An example of a nanostructured SOFC Cathode with a multiple length scale structure critical to its performance is given to emphasise the challenges in ab initio materials design (4).

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Functional electronic inversion layers at ferroelectric domain walls

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Ferroelectric domain walls hold great promise as functional two-dimensional materials because of their unusual electronic properties. Particularly intriguing are the so-called charged walls where a polarity mismatch causes local, diverging electrostatic potentials requiring charge compensation and hence a change in the electronic structure. These walls can exhibit significantly enhanced conductivity and serve as a circuit path. The development of all-domain-wall devices, however, also requires walls with controllable output to emulate electronic nano-components such as diodes and transistors. Here we demonstrate electric-field control of the electronic transport at ferroelectric domain walls. We reversibly switch from resistive to conductive behavior at charged walls in semiconducting ErMnO_3 . We relate the transition to the formation—and eventual activation—of an inversion layer that acts as the channel for the charge transport. The findings provide new insight into the domain-wall physics in ferroelectrics and foreshadow the possibility to design elementary digital devices for all-domain-wall circuitry.

Néel skyrmion lattice stabilized in iridate-manganite heterostructures

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Recent studies in the field of 3d-5d oxide heterostructures have revealed interesting physical properties at interfaces between iridates and manganites. Examples include the emergence of ferromagnetic order at both Ir and Mn sites originating from large interfacial charge transfer that results from the formation of molecular orbitals; the onset of a magnetism induced anomalous Hall effect; and control of the magnetic easy axis. To further explore the physics of 5d transition metal oxides with strong spin-orbit coupling, we have investigated a series of $(\text{LaMnO}_3)_m(\text{SrIrO}_3)_n$ superlattices grown on (001) SrTiO_3 substrates by pulsed laser epitaxy. From investigations of the physical properties through transport and Hall resistance (R_{xy}) measurements, we observed the anomalous Hall effect (AHE) in all samples below and near the Curie temperature (T_C). Interestingly, for samples with the sublayer thickness ranging $4 \leq (m, n) \leq 6$, we found a strong deviation from the standard AHE behavior at high magnetic fields (see Fig. 1). This deviation was found to arise as a consequence of the topological Hall effect owing to the presence of skyrmions. Through both density functional theory calculations and simulations of the micro magnetic structure through the object oriented micro magnetic framework, we conclude that the observation of the topological Hall effect originates from the formation of a lattice of Néel skyrmions. Note that the skyrmions form in the LaMnO_3 layers owing to the exchange coupling in the ferromagnetic insulator LaMnO_3 and a large Dzyaloshinskii-Moriya interaction (DMI) at the interface with SrIrO_3 . Note that SrIrO_3 plays a critical role in providing a large spin-orbit interaction at the interface, which greatly enhances not only the DMI, but also the interfacial exchange coupling. Importantly, the skyrmion lattice penetrates deep into SrIrO_3 layers within superlattices. Note that skyrmions observed in our system are stable in a wider range of phase space than commonly observed in metal/alloy systems. Thus, our unprecedented observation of the robust skyrmion lattice demonstrates the great potential of SrIrO_3 for developing novel quantum materials and spintronic devices.

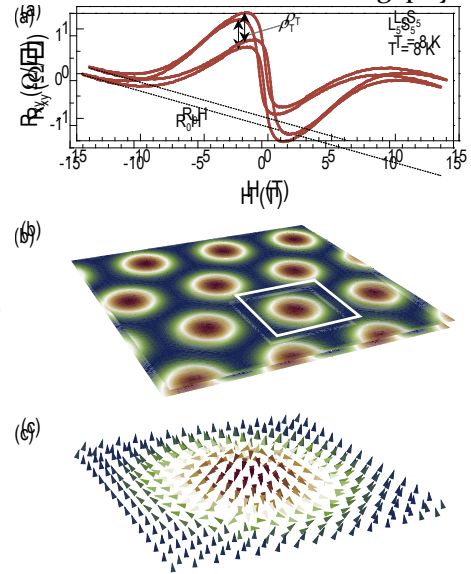


Fig. 1 a) Hall resistance of a $(\text{LaMnO}_3)_5/(\text{SrIrO}_3)_5$ superlattice measured at $T = 8$ K. b) A micro magnetic structure (image size = 18×18 nm²) from density functional theory calculations. Contour plot of component of magnetization parallel to H (M_z) where blue, white, and red represent M_z parallel, zero, and antiparallel to H , respectively. c) Magnified area (6×6 nm²) indicated in (b) with a white box. The arrows are unit vectors parallel to M .

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

Polar Metal Oxides: Science and Applications

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Polar metals are an unusual class of materials that possess both broken inversion symmetry and an electronic structure with a Fermi surface. While there are many metals that possess these two phenomena, very few oxides possess them. In this talk, I will present two examples, namely, LiOsO_3 , and $\text{Ca}_3\text{Ru}_2\text{O}_7$. For the first time, we have images “ferroelectric-like” domain walls in these metals. We explore the atomic structure and electronic properties of these walls, including “charged” and “uncharged” walls, through both experiments and density functional theory. Finally, we present some functional properties, including optoelectronic and multiferroic behavior.

Reversing ON/OFF resistance states in BaTiO₃ ferroelectric tunnel junctions: entangled role of polarization and ionic motion

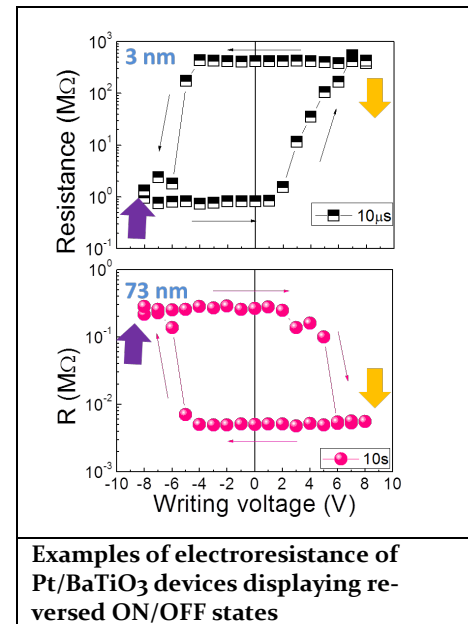
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Ferroelectric tunnel junctions (FTJs) have attracted interest for low consumption data storage applications and novel memristive devices. To win in this race, ferroelectric tunnel junctions with large and adjustable electroresistance (ER) response are required. ER is basically understood as resulting from the change of the tunnel barrier height upon polarization reversal, which produces two distinctive resistive states. In that case, the ER is expected to be a fast process. However, other mechanism may also contribute to the electroresistance, such as space-charge effects or genuine ionic motions. These processes may produce slower responses. Their combination is expected to determine and limit the switching speed between ON and OFF states, and the magnitude and sign of the electroresistance ($ER = (R_{up} - R_{down}) / R_{down}$, where R_{up} and R_{down} are the resistance values for polarizations with different orientation, dictating the ON and OFF states.

Therefore, to achieve fully understanding of its origin and exploit their properties the microscopic mechanisms contributing to ER in FTJs must be fully assessed. Here, we report on the systematic study of ER dependence on the ferroelectric barrier thickness (BaTiO₃) and writing time (τ_w) of Pt/BaTiO₃/La_{0.7}Sr_{0.3}MnO₃//SrTiO₃ junctions. Our results reveal a complex scenario where, in contrast to common wisdom [1], both the magnitude and even the sign of ER are dictated not only by electrode properties but also by the width of the ferroelectric barrier and the duration of the writing voltage pulses. For instance, we have discovered that the magnitude and sign ER can be modulated and reversed, respectively, by the duration of the writing voltage and/or temperature [2]. This finding provides clue evidences on the entangled effects of polarization switching (and the concomitant changes of tunnel barrier width and Schottky barrier height) and voltage-controlled ionic motions, on the ER of ferroelectric junctions. These observations indicate the ER is not solely dictated by the ferroelectric polarization and, may be more important, the observed broad writing time-dependence offers a new toggle towards engineering memristive devices.



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Metal-Insulator Transitions at Oxide Interfaces

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Metal-insulator transitions (MITs) that are caused by strong electron correlations are the source of some of the most interesting phenomena in materials physics. They can occur either at very high carrier densities, where on-site Coulomb interactions are strong (“strongly correlated systems”), or at ultralow densities, where long-range Coulomb interactions remain unscreened. Real materials are often significantly removed from these ideal states and contain additional, complex interactions. Furthermore, disorder, which even without correlations, can also cause localization.

Two-dimensional electron gases at complex oxide interfaces have emerged as new platforms to design, control, and understand MITs caused by strong electron correlations. There are, however, significant challenges. In particular, high sheet resistances that exceed the Mott-Ioffe-Regel limit, cause systems to be insulating at most temperatures. This preempts correlation-induced phenomena. We show several examples of such transitions and present electric field gating studies of such MITs.

The main focus of the presentation is a novel MIT that is observed in the electron system at $\text{SmTiO}_3/\text{SrTiO}_3$ interfaces. This MIT is characterized by an abrupt transition at a critical temperature, below which the resistance changes by more than an order of magnitude. The temperature of the transition systematically depends on the carrier density, which is tuned from $\sim 1 \times 10^{14} \text{ cm}^{-2}$ to $3 \times 10^{14} \text{ cm}^{-2}$ by changing the SmTiO_3 thickness. Analysis of the transport properties shows non-Fermi liquid behavior and mass enhancement as the carrier density is lowered. We compare the MIT characteristics with those of MITs in other materials systems and show that they are distinctly different in several aspects. We discuss the role of long range Coulomb interactions and of the fixed charge at the polar interface.

Metal-Insulator Transitions in Freestanding NdNiO₃ Films

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Complex transition metal oxides, owing to their correlated d electrons, provide an ideal playground for manipulation of charge, spin, orbital and lattice degrees of freedom. Metal-insulator transition (MIT) displayed in rare-earth nickelates are a clear example, where the phases and ground state of the system in thin-film form can be tuned through epitaxial strain and geometric design [1,2]. Recent interest has been focused on the role of the strain field on the Ni-O-Ni bonds and the corresponding electronic structure, which play a vital role in controlling the MIT [2]. Here we report fabrication of freestanding single-crystalline NdNiO₃ membranes using a new synthesis approach [3] and examination of their electronic properties. This offers great opportunity to study the MIT in nickelate films and heterostructures in the two-dimensional limit without substrate clamping. The ability to transfer freestanding NdNiO₃ membranes onto other templates opens a door to further access the interplay between structural rotations/distortions and spin/charge orderings in this material family, for instance, by applying unprecedented levels of external strain.

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On the origin of the metal-insulator transition in rare-earth nickelates

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Rare-earth nickelates form a very intriguing series of ABO_3 perovskite oxides, which except for LaNiO_3 , exhibit on cooling a sharp metal-insulator electronic phase transition, a concurrent structural phase transition from $Pbnm$ to $P2_1/n$ symmetry and a magnetic phase transition toward an unusual E'-type antiferromagnetic spin order. Although these compounds reveal appealing for various applications, their full exploitation is still hampered by the lack of global understanding of their different phase transitions. Here, I will first assess the ability of first-principles calculations within the DFT+U formalism to describe simultaneously the structural, dynamical, electronic and magnetic properties of rare-earth nickelates and briefly discuss these properties. Then, I will address the origin of the metal-insulator transition. I will show from first-principles calculations that it corresponds in fact to a structurally triggered phase transition¹, highlighting at the same time a first concrete example of such a kind of phase transition in simple perovskites. The origin of this mechanism will be traced back in the electronic and magnetic properties. I will illustrate that these findings are not limited to nickelates but remain also relevant in the discussion of other perovskite series like ferrites and manganites.

This work was done in close collaboration with Alain Mercy, Jordan Bieder, Jorge Iñiguez, Yajun Zhang, He Xu, Marcus Schmitt and Eric Bousquet. The research has been partly supported by the PDR project Hit4FiT from F.R.S-FNRS and the ARC project AIMED from University of Liège.

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Local strain fields and point defects in epitaxial thin films of ferroic oxides

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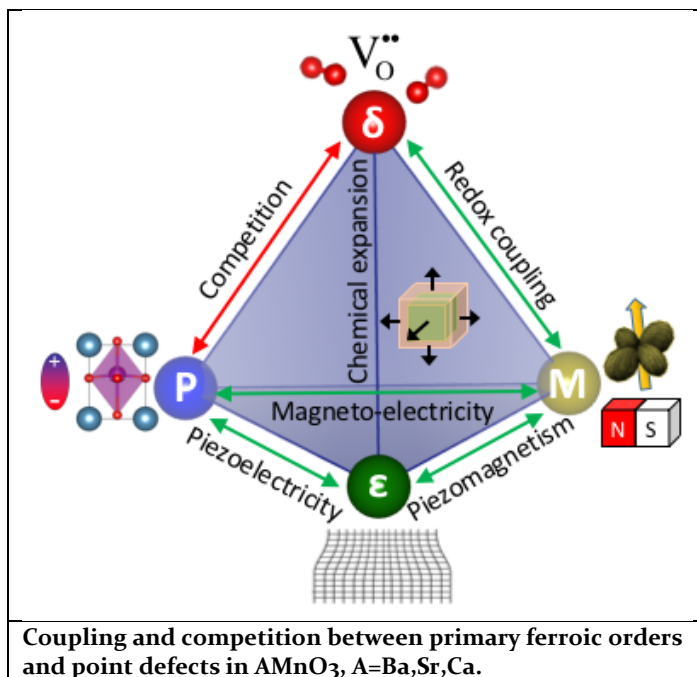
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We use density functional theory calculations to study the interplay between epitaxial strain, point defect formation and the resulting properties. The complex interplay between oxygen vacancies, A-site cation size/tolerance factor, epitaxial (001) strain, ferroelectricity, and magnetism in AMnO_3 ($A=\text{Ca}, \text{Sr}, \text{Ba}$) is illustrated in the Figure.¹ Lattice expansion by chemical pressure from large A-site cations or tensile strain favours both oxygen vacancy formation and polar distortions. Ferromagnetism is favoured by tensile strain, polar distortions and oxygen vacancies. However, polar distortions and oxygen vacancies are in a state of competition and mutual suppression. While (001) strain favours oxygen vacancy ordering, (111) strain affects all the bond lengths in octahedral equally, causing a fundamentally different strain response.

Next we address bismuth vacancy formation in BiFeO_3 under (111) and (001) epitaxial strain. While bismuth vacancy formation is generally favoured under compressive strain, the formation of $V_{\text{Bi}}\text{-}V_{\text{O}}$ vacancy pairs, is strongly favoured by epitaxial strain, implying that it is challenging to avoid local bismuth deficiency in highly strained BiFeO_3 thin films.

Finally, we study interactions between point defects and the local strain fields surrounding ferroelectric and -elastic domain walls in BiFeO_3 and improper ferroelectric YMnO_3 , and discuss implications for the processing and properties of domain walls.



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Disorder-free electrochemical doping of oxygen charge carriers into epitaxial metal-oxide films

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In material systems with strongly correlated electrons, exotic physical properties can be explored by adjusting conditions like external fields, chemical composition, doping, etc. Perhaps the most celebrated phase diagram generated by such tuning is that of the superconducting copper oxides, where introducing charge carriers into an antiferromagnetic Mott insulator renders unconventional superconductivity and a host of other fascinating phenomena. However, doping oxide materials by chemical substitution or by variation of the oxygen stoichiometry usually comes at the expense of significant disorder.

Epitaxial strain, interfacial geometries and reduced dimensionality are conditions that could yield a new generation of properties and functionalities, particularly in the case of transition metal oxides. As we improve our capabilities of growing reduced disorder thin films, multilayers and superlattices, controlled methods of carrier-doping these novel materials remain limited and relatively unexplored.

In this talk, I will discuss the development of a method to introduce oxygen into epitaxially grown, ultra-thin films of La_2CuO_4 using an *ex-situ* electrochemical technique previously employed in bulk single crystals, which yields high superconducting transition temperatures. Depending on the pH level of the electrolyte solution and aided by the substrate strain, the dopants self-arrange into unusual periodicities with extremely long-range coherence. Thus, we identify a new and useful way of doping epitaxial heterostructures of transition metal oxides with remarkably low disorder.

Defect Mitigating $(\text{SrTiO}_3)_n(\text{BaTiO}_3)_m\text{SrO}$ Superlattices for mmWave Tunable Dielectrics

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The Ruddlesden-Popper (RP) superlattice series, $(\text{ABO}_3)_n\text{AO}$, has been identified as a defect mitigating structure via a proposed mechanism in which the $(\text{AO})_2$ layer accommodates the local non-stoichiometry of the crystal by changing its area. The accommodation of the $(\text{AO})_2$ layers allows the perovskite $(\text{ABO}_3)_n$ sections of the material to remain stoichiometric. In the strained $(\text{SrTiO}_3)_n\text{SrO}$ thin film phase we recently demonstrated record tunable dielectric performance at gigahertz (mmWave) frequencies, a region where point defects significantly contribute to dielectric loss. In contrast, the parent phase commonly used for frequency tunable microwave circuit elements, $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$, experiences large attenuation at gigahertz frequencies arising from dielectric loss related to point defects. We use oxide molecular-beam epitaxy (MBE) as a controlled way to fabricate the next generation of thin-film, mmWave tunable dielectrics via precise atomic layering and epitaxial strain. In this work we use MBE to grow related RP phases containing $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$, specifically $(\text{SrTiO}_3)_n(\text{BaTiO}_3)_m\text{SrO}$ shown in figure 1, in hopes of accommodating point defects by the $(\text{SrO})_2$ faults, allowing the rest of the dielectric material to remain stoichiometric.

An advantage of these Ba-containing RP phases is lower epitaxial strain due to BaTiO_3 's smaller lattice mismatch with the DyScO_3 (110) substrate, which allows for thicker films without the loss of the desired low-temperature ferroelectric instability. The first five members of this RP homologous series ($n = 1-5$) have been grown on DyScO_3 (110) using MBE and characterized by x-ray diffraction. We use density functional theory (DFT) to predict and understand the ferroelectric properties of these films. In-plane measurements of the dielectric constant have been made as a function of temperature and frequency in the gigahertz frequency regime. We use

scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) to identify barium and $(\text{SrO})_2$ fault placement, and find high quality films with 1-3 atomic layers of barium interdiffusion, despite the metastability of these atomically engineered structures.

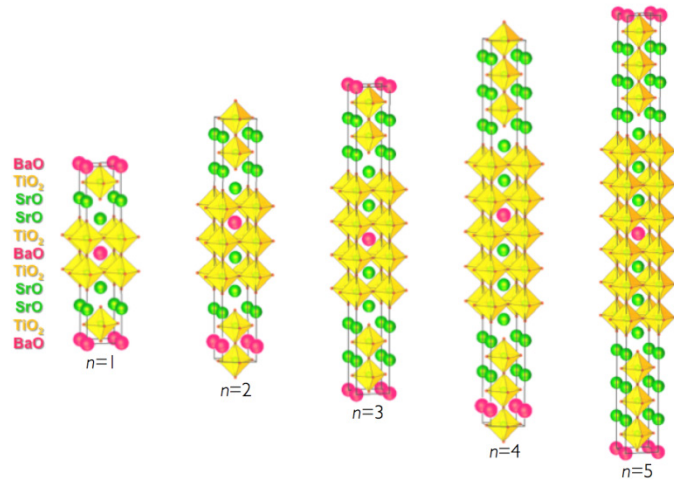


Figure 1. Schematic of the unit cell crystal structure of $(\text{SrTiO}_3)_n(\text{BaTiO}_3)_m\text{SrO}$ Ruddlesden-Popper phases for $m=1$, $n=1-5$.

Enhanced lithium transport in highly ordered LiMn_2O_4 cathode films towards solid-state batteries

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Solid-state microbatteries can facilitate miniaturization, create more flexibility for the design of stand-alone microelectronic devices and enhance their applicability, for example in medical implants, due to the avoided leakage risks. However, the successful application of all-solid-state microbatteries depends strongly on the enhancement of energy density and lifetime. The cycle-life and lifetime are dependent on the nature of the interfaces between the electrodes and electrolyte, whereas safety is a function of the stability of the electrode materials and interfaces. Therefore, perfect control on the interfacial properties between the electrodes and electrolyte is needed, but remains a great challenge.

Various oxide materials are known as promising cathode materials as they provide good thermochemical stability, high energy density and high voltage. However, the crystal structure of the main candidates (spinel LiMn_2O_4 , layered LiCoO_2 and olivine LiFePO_4) exhibit distinct differences in the lithium diffusion behavior along 3D, 2D and 1D directions respectively. Common bulk studies on battery materials investigate polycrystalline films and, therefore, do not enable detailed characterization of the ionic and electrical conductivity along specific crystalline orientations. Furthermore, the formation of the unwanted solid electrode interphase (SEI) layer between a cathode and electrolyte varies strongly depending on the specific crystal facet of the cathode material. Single crystalline thin films of cathode materials are required to obtain more insight into the ideal interface ordering and the corresponding ionic and electrical properties.

Here, we have studied the lithium diffusion in LiMn_2O_4 thin films, which are epitaxially grown by pulsed laser deposition on single crystalline Nb-doped SrTiO_3 substrates. Control over the specific crystal orientation of the full thin film enables detailed analysis of the lithium diffusion along specific crystal planes ($\{001\}$, $\{110\}$ and $\{111\}$). Single phase LiMn_2O_4 films show enhanced cyclability and faster charging speed, as compared to studies on polycrystalline materials. The achieved capacity reached >90% of the theoretical limit and no capacity reduction was observed after the first cycle when measured over 300 cycles. High charge-discharge speeds in the range 1C-10C showed good capacity retention.

To achieve insight into the non-uniform distribution of lithium activity at the cathode-electrolyte interface, electrochemical strain microscopy (ESM) is applied. This enables nanoscale mapping of intercalation, strain and charge & ion transport at the level of single grain boundaries and dislocations down to sub-20 nm scale. The local electrochemical reaction and transport processes are decoupled and probed independently by adding spectroscopic 2D first-order reversal curve (FORC) measurements. This technique offers the capability to distinguish local regions with low effective mobility of lithium ions from other regions with high lithium mobility. Additionally, voltage pulses of increasing and decreasing amplitude are applied to determine the local dynamic voltage-activated electrochemical processes and transport.

Interaction of CO₂ and NO_x gas on transition metal perovskites

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Transition metal perovskite oxides (ABO₃, A = rare earth, B = transition metal) provide a rich and flexible platform for bulk electronic and chemical design.¹ As a result, they have been employed extensively as catalysts to enable clean air - NO_x removal^{2,3} and volatile organic compound oxidation⁴ – and electrochemical energy conversion processes - metal-air batteries⁵, (photo)catalytic water-splitting or CO₂ conversion⁶, and oxygen-based fuel cells⁷. Understanding and designing metal oxide surface chemistry in these systems is critical for the continued development of stable and active catalysts for these processes.

In this study, we present two case studies of CO₂ gas and NO_x mixtures interaction with cobalt perovskite chemistries. By employing near ambient pressure x-ray photoelectron spectroscopy (AP-XPS), the gas-surface interface can be studied at previously inaccessible thermodynamic states by surface science techniques.⁸ In combination with epitaxial thin films with well-defined surfaces, this enables the quantification of species formation and their systematic comparison among chemistries. We find that the bulk oxygen 2p band center relative to the Fermi level governs the speciation of carbonates and NO reactivity in CO₂ and NO_x gas environments, respectively. These observations can be rationalized by the nature of the M-O covalent bond and oxygen vacancy formation, as determined by the O 2p band level.⁹ We highlight the relevance of these findings for perovskite surface chemistries involving carbon chemistries and NO_x removal reactions.

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2D electron system at the magnetically tunable EuO/SrTiO₃ interface

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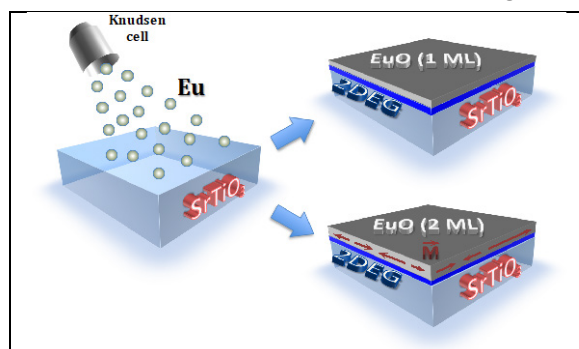
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Two-dimensional electron systems (2DESs) in functional oxides have gained strong interest as a novel state of matter with fascinating and exotic interface physics. The prospect of creating and manipulating a macroscopic magnetic ground state in oxide-based 2DESs is of enormous interest, as this would pave the route towards oxide spintronic applications with novel quantum phases beyond today's semiconductor technology.

We create a 2DES with magnetic functionalities at the interface between EuO, a ferromagnetic insulator, and SrTiO₃ - a transparent non-magnetic insulator considered the bedrock of oxide-based electronics. This is achieved by a controlled *in situ* redox reaction between pure metallic Eu deposited at room temperature on the surface of SrTiO₃ - an innovative bottom-up approach that can be easily generalized to other functional oxides and scaled to applications.

Additionally, we find that the resulting EuO capping layer can be tuned from paramagnetic to ferromagnetic, depending on the Eu metal coverage (1 ML and 2 ML, respectively), and show, using angle-resolved photoemission spectroscopy (ARPES), that the integrity of the 2DES is preserved in both cases, thus providing an ideal knob for tuning the spin-transport properties of the 2DES. These results demonstrate that the simple, novel technique of creating 2DESs in oxides by deposition of elementary reducing agents [1] can be extended to simultaneously produce an active, e.g. magnetic, capping layer enabling the realization and control of additional functionalities in such oxide-based 2DESs. The thickness-controlled tuning of the overlayer magnetic properties [2] opens exciting possibilities for the fabrication of nano-circuits coupling magnetically-gated 2DESs to “standard” 2DESs in oxide systems. These results lay a new ground for the simple and versatile design of all-oxide devices in which the functionalities of the constituting elements, and their mutual coupling, can be obtained from controlled physicochemical reactions and vacancy engineering at their interfaces.



Schematics of the experiment. Pure Eu metal reacts with the SrTiO₃ surface, forming stoichiometric insulating EuO. The redox reaction locally reduces the SrTiO₃ around its surface, creating a 2DEG (blue). The capping layer can be tuned from paramagnetic, for 1 ML of EuO, to ferromagnetic, for 2 ML of EuO.

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Control of Ferromagnetism in Ion-Gel-Gated $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ Thin Films

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Recently, electrolyte gating techniques employing ionic liquids have proven remarkably effective in tuning large carrier densities at the surface of a variety of materials. These include organic conductors, semiconductors, 2D materials, binary oxides, transition metal oxides, perovskites, and other complex oxides. In essence these electrolytes enable electric double layer transistor operation, the large specific capacitances (10's of $\mu\text{F}/\text{cm}^2$) generating electron/hole densities up to 10^{14} - 10^{15} cm^{-2} , *i.e.*, significant fractions of an electron/hole per unit cell in most materials. Successes include discovery of superconductivity in KTaO_3 , tuning over a dome of superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, and electrical control of the insulator-metal transition in VO_2 , although open questions remain. Uncertainties include the true doping mechanism (*i.e.*, electrostatic vs. electrochemical (*e.g.*, redox-based)), the relation between 2D surface and bulk chemical doping, the role of electrostatic disorder, the need for *in operando* characterization, and the universality of the approach. In this talk we will review the application of electrolyte gating using not ionic liquids, but ionic *gels* (or *ion gels*), which enable simple processing of all-solid-state devices. [1-3] We focus on ultrathin epitaxial films of the perovskite $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$, seeking gate-control of ferromagnetism. Our findings first clarify the issue of charge carrier vs. oxygen defect creation, transport measurements revealing a dramatic asymmetry with respect to bias polarity, with important implications for *n*-type oxides such as SrTiO_3 and VO_2 . [4] We then report on the development of *in operando* probes of ion-gel/ $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ devices, using hard X-ray synchrotron diffraction and Polarized Neutron Reflectometry (PNR). These provide direct evidence of oxygen vacancy creation at positive bias, vs. electrostatic hole accumulation at negative bias. Gate-control over resistivity, magnetoresistance, magnetization, and Curie temperature is thus demonstrated under hole accumulation, including gate-induced ferromagnetism directly verified by *in situ* PNR. These results are discussed in terms of recently developed theory for electrolyte-gate-induced percolation [5].

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A New Computing Substrate Enabled by Functional Oxides

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While deterministic Boolean logic has been the backbone of information processing, there are computationally hard problems like solving constrained optimization problems wherein this traditional paradigm is fundamentally inadequate. This results in computational inefficacy, and motivates us to explore new pathways to their solution. In this invited talk, we introduce an experimental testbed comprising of compact coupled relaxation oscillator based dynamical system that exploits the insulator-metal transition in correlated oxide exhibiting insulator-to-metal phase transition (for example VO₂), to efficiently solve the vertex coloring of arbitrary graphs, a prototypical combinatorial optimization problem. Our work is inspired by the understanding that optimization finds a natural analogue in the energy minimization process of highly parallel, coupled dynamical systems. Our work not only elucidates a physics-based natural computing substrate that is experimentally accessible but also presents opportunities for building customized analog co-processors for solving computationally hard problems efficiently.

Designing type II neurons via instability of VO₂ metal-insulator transition

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Electronically implementing neuron function is essential for fabricating the next-generation integrated circuit, so-called neuromorphic circuit. The basic neuron function is to generate output spike signals when receiving over-threshold input, and is known to be categorized into two groups: type I and type II (Fig. 1a). The type I neuron linearly increases its output spike frequency from zero, and has widely been implemented by transistors and capacitors [1]. On the other hand, the type II neuron discontinuously changes its output spike frequency from zero to a specific value, and is known to be more critical for the organized behavior in the neuronal network [2]. However, the type II neuron needs to design instability in its operation for generating limit cycles, posing fundamental challenges for simplifying and scaling the neuromorphic circuits [2].

One of the possible solutions for this problem is to use the intrinsic instability inside the metal-insulator transition materials such as VO₂. Because its instability can generate limit cycles only with a capacitor and a resistor [3,4], it may implement the type II neuron in a considerably simple manner. In this study, we successfully demonstrate the type II neuron by using the metal-insulator transition of VO₂. The discontinuous change in the output spike frequency is achieved by switching between two limit cycles in the current-voltage plane (Fig. 1b), which are designed by combining two VO₂. Furthermore, the switching between the limit cycles is captured by oscilloscope (Fig. 1b), indicating a novel opportunity of the metal-insulator transition materials in the neuromorphic circuit. This work was supported by JST CREST Grant Number JPMJCR14F2, Japan, and was partially supported by JSPS KAKENHI 17H04812.

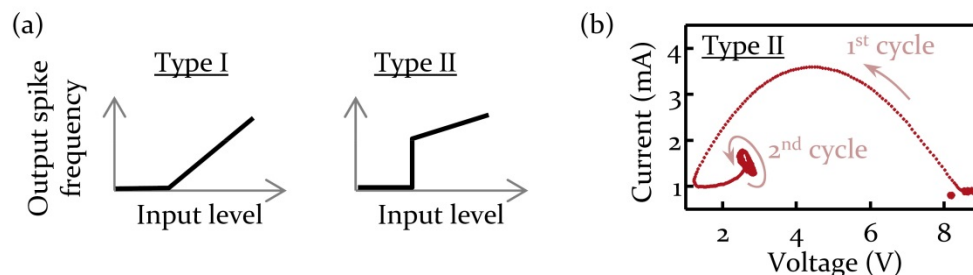


Fig. 1: (a) Two different neuron functions: type I and type II. (b) Dynamic current-voltage characteristics of the type II neuron using VO₂, measured by oscilloscope.

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3d-5d Double Perovskites: Playground for Oxide Electronics

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Double perovskites ($A_2BB'O_6$) consisting of alkaline-earth or rare-earth metal ion at A site, and two different transition metal ions at B and B' sites, are of significant interest due to the diverse properties exhibited by them. Presence of two transition metal(TM) ions instead of one as in conventional single-unit perovskites, opens up a large flexibility in choice of B site cations encompassing elements from different transition metal series. In this talk we will consider double perovskites with B site elements from 3d TM series and B' site elements from 5d TM series. Employing first principles study we will demonstrate engineering of half-metallic behavior [1] in doped Sr_2CrOsO_6 double perovskite with very high transition temperature of ~ 700 K. In the quantum well geometry of bilayer of Ba_2FeReO_6 double perovskite, sandwiched between thick layers of $BaTiO_3$, we will establish that controlled confinement of half-metallic two-dimensional electron gas in $BaTiO_3/Ba_2FeReO_6/BaTiO_3$ heterostructures is possible [2]. Our work in this respect opens up the possibility of realizing ultrathin spintronic devices. Further, considering overlayer geometry of Ba_2FeReO_6 on $BaTiO_3$ we will show that designing of high-temperature large-gap quantum anomalous Hall insulating state in ultrathin double perovskite films is achievable [3].

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***In situ* observation of layer-by-layer mean inner potential oscillations and precise growth control of oxide interfaces**

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A variety of novel quantum phases have been discovered at oxide interfaces with technical advances in deposition techniques, especially with the application of *in situ* reflection high-energy electron diffraction (RHEED) in oxide film growth. Typically, one period of the RHEED intensity oscillations corresponds to the growth of one repeat unit (e.g. an unit cell), which can be utilized to precisely control the number of repeat units in the film growth. However, the precise control of oxide surfaces/interfaces is still very challenging due to the rather complicated RHEED oscillation patterns in the oxide film growth and the lack of chemical information in the electron diffraction.

Here, we report the observation of layer-by-layer variations of the mean inner potential during the epitaxial growth of SrTiO₃ films, providing important information about the chemical composition of the surface layer during the film growth. A model including the effects of mean inner potential and surface roughness is proposed to reveal the underlying mechanism of the complicated RHEED intensity oscillations observed in the growth of oxide films. General rules are also proposed to guide the fabrication of ultimate precise oxide surfaces/interfaces for novel correlated quantum states. As an example, atomic sharp LaAlO₃/SrTiO₃ interfaces and two dimensional electron gas (2DEG) were successfully fabricated on epitaxial SrTiO₃ films of any desired thicknesses, shedding light on the *in situ* fabrication of oxide 2DEG and other novel interfacial phases on preferred templates but not limited to the pre-etched TiO₂-terminated SrTiO₃ substrates.

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Suppressed disorder in superconducting $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ epitaxial thin films

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Recent predictions of a wide-gap topological insulating state in BaBiO_3 has stimulated significant interest, in part because BaBiO_3 hosts superconductivity with Pb or K doping.¹ The simultaneous existence of these two quantum phases in an oxide is ideal for synthesizing heterostructures that enable study of Majorana physics or engineering new high temperature superconducting phases. However, few reports exist demonstrating the high quality typical in other oxide epitaxial films. Here we demonstrate that the large lattice constant of BaBiO_3 materials ($a_{\text{pc}} \approx 4.3 \text{ \AA}$) severely limits material quality and properties in thin $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ (BPBO) superconducting films when grown on the commonly used SrTiO_3 substrate ($a_{\text{pc}} \approx 3.9 \text{ \AA}$). Thick BPBO films exhibit bulk like transition temperature ($\sim 11 \text{ K}$) and the lowest reported resistivity; however, transition temperatures are significantly depressed with decreasing thickness, consistent with a disorder induced superconductor-insulator transition. By moving to LaLuO_3 substrates ($a_{\text{pc}} \approx 4.18 \text{ \AA}$) with better lattice matching, thin BPBO exhibit higher superconducting transition temperatures, smoother surfaces, and higher crystal quality. The sensitivity of BPBO to disorder caused by poor lattice matching is an important consideration as the community attempts to design new properties using thin heterostructures.

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Control of oxygen coordination environment in transition-metal oxides

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Oxygen coordination environments of transition metals strongly influence the properties of oxide materials. Two recent topics of controlling the oxygen coordination environments are highlighted in the talk.

The first topic concerns topochemical oxidation/reduction of transition-metal oxides. The oxidation states of transition metals can be changed by modifying the oxygen coordination, and unusual oxidation states can often be stabilized. To relieve the electronic instabilities of such unusual oxidation states of the transition-metal cations, charge transitions are induced and drastic changes in physical properties are often observed. The intriguing charge behaviors found in some transition-metal oxides with unusual high-valence Fe ions are reviewed [1-4].

The second topic focuses on the arrangement of oxygen ions at heterointerfaces consisting of dissimilar oxides. We have found significant modification of oxygen coordination environments at the interface between $\text{SrFeO}_{2.5}$ and DyScO_3 . When the brownmillerite-structure $\text{SrFeO}_{2.5}$ film grows epitaxially on a perovskite DyScO_3 substrate, the alternately ordered arrangement of oxygen is significantly disturbed and reconstructed in the heterointerface region to accommodate structural mismatch [5]. The unusual oxygen coordination environment that otherwise is not stable at ambient conditions will be discussed.

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Unusual orbitally-polarized insulating state at a titanate/cobaltate interface

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Advanced oxide MBE growth is able to fabricate artificial layered oxide materials where the cation composition of each 2D layer can be controlled at will. When the 2D layers themselves are only a unit cell thick, interfacial phenomena move the forefront. We describe first principles theoretical results, that go hand in hand with parallel experimental studies, on an unusual titanate/cobaltate charge-transfer interface that is formed in (001) $(\text{LaCoO}_3)_2/(\text{LaTiO}_3)_2$ superlattices. Electrons move across the interface from Ti t_{2g} states to Co e_g states, and the resulting physical and electronic structure leads to a strong orbital polarization within the Co e_g manifold. The resulting electronic structure is rich and includes a number of insulating solutions with close energies: these include magnetically ordered states as well as an unexpected paramagnetic insulating state where Co $d_{3z^2-r^2}$ orbitals along (001) pair up to form a filled molecular dimer states, a realization of the “Dimer Mott” state [1]. We will describe the fascinating interplay between electron transfer, polar distortions, orbital polarization, and magnetic structure at this unique interface.

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Growth of antiperovskite-type oxide Ca_3SnO thin films by pulsed laser deposition

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Perovskite oxides and their heterostructures are well known as one of the valuable systems showing novel functional properties. Recently, their counterparts, namely antiperovskite oxides A_3BO have been attracting much attention as an alternative platform for searching unique physical properties. Immediately after the theoretical prediction that some of the A_3BO family have three-dimensional (3D) Dirac Fermions [1], a number of experimental and theoretical efforts has been devoted to exploring possible bulk Dirac Fermions in A_3BO [2,3]. Since this unique electronic structure appears not at the surface but in the bulk, the heterostructures composed of perovskite and antiperovskite might also be promising mine to explore novel physical properties in the light of the analogies to the perovskite oxide heterostructures. However, despite these intriguing research subjects, the study on the growth of antiperovskite films is limited due to its unusual anionic state and air sensitivity [2,3].

In this study, we report the epitaxial growth of antiperovskite Ca_3SnO films on (001)-oriented cubic yttria-stabilized zirconia (YSZ) substrates by using pulsed laser deposition (PLD) technique. Unlike the previous studies [2], sintered Ca_3SnO pellet was used as the ablation target. By tuning the growth conditions, we have succeeded in growing the epitaxial films of Ca_3SnO . The achievement of its flat surface was confirmed by the streak pattern in reflection high-energy electron diffraction patterns. The epitaxial nature of the films on YSZ substrates was proved by x-ray diffraction. The Ca_3SnO films were epitaxially grown on YSZ substrates with the cube-on-cube orientation relationship, although the existence of some impurity phases was surveyed. The electronic structures, as well as the chemical composition, of the grown films were examined by x-ray photoemission spectroscopy. The shape of valence band spectra was qualitatively reproduced by the first principles calculation, although there were discrepancies between the two near the Fermi level. These results strongly suggest that antiperovskite films is directly synthesized from the target by using a PLD technique.

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In-plane spin cycloid structure in multiferroic BiFeO₃ thin films

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Widely hailed as one of the first room-temperature functional multiferroic materials, BiFeO₃ exhibits coexisting ferroelectric ($T_C \sim 1103$ K) and antiferromagnetic ($T_N \sim 643$ K) order [1]. Although the spontaneous electric polarization in the bulk phase is a modest $6.1 \mu\text{C}/\text{cm}^2$, it can be increased to as much as $102 \mu\text{C}/\text{cm}^2$ in BiFeO₃ that has been stabilized as an epitaxial thin film [2]. This enhancement has been attributed to strain engineering, but also to the suppression of a long range cycloidal modulation of the antiferromagnetic spin structure that exists in bulk crystals [3].

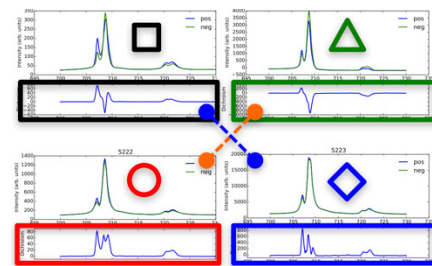
Recent measurements of the spin structure in BiFeO₃ films have challenged this assertion. It has been suggested that various spin cycloids can exist in strained BiFeO₃ films that are sufficiently thick to support the ~ 60 -nm modulations [4].

We have systematically investigated the magnetic ordering in BiFeO₃ films using resonant soft x-ray diffraction (RSXD). Films were grown on DyScO₃ (110)₀ to create periodically alternating domain stripes with 71-degree walls, as confirmed by piezoelectric force microscopy. RSXD with circularly polarized x-rays tuned to the iron $L_{3,2}$ transition edges revealed a long-range magnetic modulation in these films. Symmetry analysis of the polarization-dependent diffraction peaks indicates that a spin cycloid with in-plane periodicity exists in BiFeO₃ films down to ~ 10 -nm thickness. These results have significant implications for inter-/intra-domain magnetoelectric coupling, as well as magnetic coupling between BiFeO₃ and other magnetic materials.

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The complex symmetry of circular dichroism in RSXD peaks reveal the in-plane spin cycloid in BiFeO₃ films.

Selective control of multiple ferroelectric switching pathways using trailing flexoelectric field

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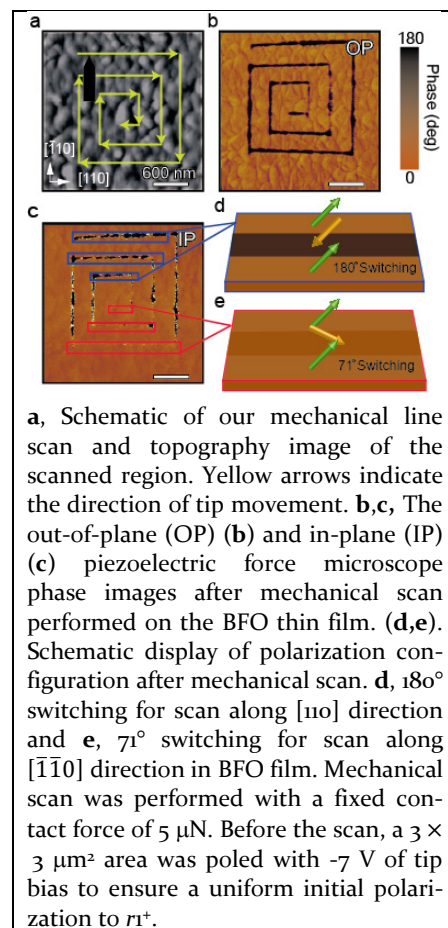
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While conventionally controlled by the electrical bias of a scanning probe, the polarization domain in ferroelectric thin films can also be reoriented by mechanical force through the tip by virtue of the flexoelectric effect. Although the mechanical approach for polarization switching has been demonstrated in a number of ferroelectric materials, most investigations concern only the 180° reversal of the out-of-plane polarization¹. However, a systematic study is still lacking for the domain switching in more complex situations where both non- 180° ferroelastic and 180° ferroelectric switching coexist.

In ferroelectric materials, more than one polarization switching pathway can be an important ingredient for novel electronic devices, such as multilevel memory storage. Especially, if the ferroelectric materials possess other ferroic ordering, namely like multiferroic BiFeO_3 (BFO), the multiple switching pathways can be utilized for magnetoelectric devices. However, for actual realization of those devices, deterministic control of multiple polarization switching pathways is an essential prerequisite. Although there were many efforts to selectively control the ferroelectric switching pathways and several studies have shown its possibility, typically they required additional laborious processes^{2,3}.

In our work, we show that as a new non-electrical approach, 'trailing flexoelectric field' offers a simple but very effective route for selective control of multiple ferroelectric switching pathways in a BFO thin film. The simple pressing of the electrically grounded SPM tip and the control of its motion allow us to select particular domain switching only.



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Topological functions in oxide heterostructures

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Recent advancement in the research of topological quantum materials enables us to think about dissipation-less electronics based on topological current at the edge. The origin of this field is the discovery of quantum Hall effect (QHE) in two dimensional electron system. Above room temperature QHE has been achieved in a Dirac system of graphene, when chemical potential is set within the cyclotron gap. Zero magnetic field QHE has been realized in a magnetically doped topological insulators, where chemical potential is set within the mass gap generated in a Dirac system by broken time-reversal symmetry. We are paying effort to extend this trend in oxide world.

One such example is possible Weyl semimetal in pyrochlore Ir oxides. Peculiar all-in-all-out antiferromagnetic spin texture gives unprecedented magnetoresistance with odd-parity. The domain boundary is theoretically predicted to be metal due to topological band inversion, which is experimentally verified.

Another possible example is the high mobility electron system in ferromagnetic semiconductor EuTiO₃. In addition to the ordinary Hall effect being proportional to magnetic field, it shows anomalous Hall effect being proportional to magnetization and topological Hall effect. The origin of latter is still unclear but it may be a signature of non-trivial spin texture like skyrmion that has finite scalar spin chirality. Other examples are also presented for skyrmion hosted in oxide heterostructures.

Unusual magnetism in LaNiO₃-based superlattices

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LaNiO₃ is the only member of the perovskite nickelates which in bulk remains metallic and paramagnetic at all temperatures. However, a transition to an insulating state occurs in thin films as its thickness is reduced to few unit cells.¹ We show here that antiferromagnetism can also be stabilized in ultrathin insulating LaNiO₃ layers when grown along the (111)-direction in LaNiO₃/LaMnO₃ superlattices. As a result, negative exchange bias is observed at low temperatures.² For the case of superlattices with 7-monolayers-thick-LaNiO₃, an evolution from negative to positive exchange bias behaviour is observed as temperature is increased, followed by an antiferromagnetic interlayer-coupled state between LaMnO₃ layers above the blocking temperature.³ The emergent antiferromagnetic order in LaNiO₃ is described by a $(1/4, 1/4, 1/4)_{\text{pc}}$ wavevector, akin to the one displayed by all the other nickelates in the insulating phase. The reduced dimensionalities of the nickelate layer and interfacial charge transfer are key to stabilize such a magnetic state. Moreover, the presence of a structural asymmetry at the LaNiO₃-LaMnO₃ interfaces as revealed by our TEM studies⁴ is also required to explain the complex magnetic evolution of the system. To further investigate the behaviour of these heterostructures, tricolour superlattices of the type LaNiO₃/LaMnO₃/LaAlO₃ will also be discussed.

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Controlling the magnetic spin reconstruction in (111)-oriented $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/\text{LaFeO}_3$ heterostructures

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Transition metal oxides exhibit strong coupling between atomic structure and magnetic properties, enabling us to engineer epitaxial heterostructures with emerging magnetic properties at epitaxial interfaces. (Bhattacharya and May 2014) Here, we present a study of controlling the induced ferromagnetic moment of Fe by imposing different structural symmetries using geometrical lattice engineering. As a model system we investigate epitaxial heterostructures consisting of antiferromagnetic LaFeO_3 (LFO) and ferromagnetic $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) grown in the (111)-interface orientation. By relying on this crystalline facet the structural coupling between the two is large hence allowing to maximize possible magnetic reconstructions. For quadratic strain by having SrTiO_3 as substrate an induced ferromagnetic moment, $\sim 1.6\text{--}2.0 \mu_B/\text{Fe atom}$, is found antiparallel to the ferromagnetic moments of LSMO for LFO thicknesses less than 4 nm. (Hallsteinsen, Moreau et al. 2016) This effect spans 2–4 monolayers in the LFO from the LSMO interface, while the rest of the LFO is antiferromagnetically ordered with an out-of-plane component of the antiferromagnetic axis. The interplay between structural domains in LaFeO_3 , oxygen octahedral reconstructions at the interface, the antiferromagnetic LFO spin structure and the magnetic state of LSMO affects the observed switchable Fe-moment. To decouple the effects, we study how the magnetic configuration and the induced moment change in LSMO/LFO heterostructures with interfaces oriented in different crystallographic directions, with varying layer thicknesses and deposited on different substrates inducing different strain states. We will especially study the symmetry of LFO, dependent on the substrate symmetry, in order to probe the role of interface octahedral structure on the magnetic properties. The magnetic and atomic structure is characterized using a combination of x-ray spectroscopy, neutron reflectometry and electron microscopy.

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Tuning the properties of oxide heterostructures by interfacial oxygen octahedral coupling

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Diverse electronic phases in solid state materials such as superconductivity, topological insulating phases, ferroelectricity and ferromagnetism are intimately coupled to the crystal symmetry. In ABO_3 perovskites, the crystal symmetry resides in the corner sharing oxygen octahedral (BO_6) network. These symmetries, or oxygen octahedral rotation (OOR) patterns in perovskite heterostructures, are usually engineered by epitaxial strain. Furthermore, the required connectivity of the octahedra across the heterostructure interface enforces a geometric constraint to the 3-dimensional octahedral network in epitaxial films. Such geometric constraint will either change the tilt angle to retain the connectivity of the corner shared oxygen octahedral network or guide the formation of a specific symmetry throughout the epitaxial film. Although the impact of the oxygen octahedral coupling (OOC) on the tilt angle is found to be limited to a short range of only 4-8 unit cells (uc), the OOC-induced symmetry can propagate throughout the film. Therefore, control of the OOC provides a large degree of freedom to manipulate physical phenomena in complex oxide heterostructures.

In my presentation, I will discuss the control of oxygen octahedral coupling by interface-engineering in manganite as well as nickelate heterostructures. Control of the oxygen octahedral coupling will be discussed, which enables tuning of the lateral magnetic and electrical anisotropy in manganite heterostructures and the metal-insulator transition (MIT) in nickelate heterostructures. Due to the interfacial octahedral coupling effect we are able to manipulate the tilt angle of the SmNiO_3 octahedra and in turn successfully control the MIT. The lattice structure across the superlattices, as revealed by scanning transmission electron microscopy (STEM), atomically links a reduced MIT temperature to a smaller SmNiO_6 tilt angles. X-ray absorption spectroscopy and resonance magnetic X-ray diffraction further reveal a central role of the Ni-O-Ni bond angle in determining the electronic ground state of the nickelate material. Such design of the octahedral configuration in oxide heterostructures provides us a powerful strategy to manipulate the transport properties and to achieve desired functionalities in correlated oxides.

Manipulating $\text{LaInO}_3/\text{BaSnO}_3$ polar interface

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We have recently reported on the conducting $\text{LaInO}_3/\text{BaSnO}_3$ polar interface. [1] We have found that a small amount of doping in the BaSnO_3 layer is critical for the conducting interface behavior, while the interface between the LaInO_3 and the undoped BaSnO_3 was not conducting. By way of series of experiments, we show that the conducting behavior is due to neither oxygen vacancies nor cation diffusion at the interface. One of such experiments is a modulation doped heterostructure, where an undoped BaSnO_3 spacer layer is inserted between the LaInO_3 and the slightly doped BaSnO_3 . [2] We will report on the electrical properties of such structures. We will also report on the properties of the BaSnO_3 interface with other larger bandgap perovskite materials, including nonpolar BaHfO_3 and SrZrO_3 epitaxial perovskites. Another approach to manipulate the $\text{LaInO}_3/\text{BaSnO}_3$ interface is to modify the epitaxial strain at the interface as an attempt to change the interface polarization responsible for the 2DEG behavior. We will present our efforts to correlate the electrical properties with the structural properties of such interfaces.

Our model for the conducting interface is based on the 2DEG-like structure created by interface polarization of the LaInO_3 and the Fermi level controlled by the doping in the BaSnO_3 . Using such $\text{LaInO}_3/\text{BaSnO}_3$ conducting interfaces, all perovskite transparent FETs with excellent properties were made. [3] We believe that the stable oxygen stoichiometry and the ability to control the local doping level in $\text{LaInO}_3/\text{BaSnO}_3$ heterostructures will lead to better understanding of the 2DEG behavior, tuning and creating higher density 2DEG, and eventually further increasing the mobility.

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Surface and Interfacial Phenomena in the $\text{LaFeO}_3/n\text{-SrTiO}_3$ Heterojunction

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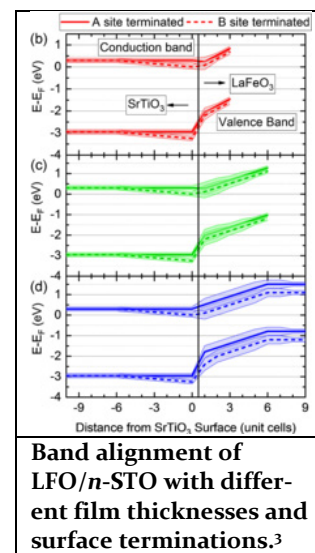
Polar/non-polar interfaces in epitaxial oxide films have been a rich area of research for many years for emergent behavior. The interfacial dipole that results from the polar discontinuity at such interfaces can generate electronic reconstruction near the interface, which may be used to separate optically-excited electron-hole pairs for enhanced photovoltaic and photocatalytic response. Recent studies of the LaFeO_3 (LFO)/Nb-doped SrTiO_3 ($n\text{-STO}$) interface have shown exciting photoconductive and photocatalytic behavior.^{1,2} In these studies, interfacial polarity (i.e. positively charged $\text{TiO}_2/\text{LaO}^+$ or negatively charged $\text{SrO}/\text{FeO}_2^-$) was shown to affect the photoresponse of the materials.

Here we present studies of LFO/ $n\text{-STO}$ epitaxial interfaces with differing interfacial polarity for films grown by molecular beam epitaxy. Films are grown layer-by-layer on TiO_2 and SrO-terminated $n\text{-STO}$ substrates to carefully engineer the interfacial and surface terminations. Using *in situ* x-ray photoelectron spectroscopy measurements, we estimate the valence and conduction band electronic alignment on both sides of the interface.³ These results are correlated with scanning transmission electron microscopy measurements and density functional theory modeling of the interface to understand the behavior of the positively and negatively charged interfaces during growth. We also present ambient-pressure XPS studies of these samples to show that the surface-termination strongly impacts the reactivity of the samples with water for their use in photocatalytic applications.⁴

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Revealing topological states at a hybrid oxide chalcogenide interface

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The development of chemically and electrically robust topological surface states in topological insulators (TIs) and topological crystalline insulators (TCIs) is a critical step toward incorporating their unique properties, such as spin-momentum locking, into functional electronic devices. We have achieved controlled ultra-thin film growth of TCI SnTe on SrTiO₃ by adapting conventional molecular beam epitaxy methods. Thin SnTe films (6-25 nm-thick) are uniform and have high crystalline quality. Resistivity measurements show suppression of bulk conduction in thinner films while maintaining conduction through surface states. By measuring the Fermi level at the surface and band alignments of SnTe and SrTiO₃, we determine that quantum transport of a topological nature occurs at the buried SnTe/SrTiO₃ interface.

Multifunctional electronic states of 2DEGs at perovskite oxide surfaces

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Following the 2011 experimental report of a two-dimensional electron fluid (2DEG) at the bare surface of SrTiO₃, a variety of insulating transition metal oxide perovskites (e.g. KTaO₃, BaTiO₃, CaTiO₃, anatase TiO₂) have been shown to exhibit similar states at their surface, for various types of orientations. Orbital reconstruction as well as oxygen vacancies at the surface plays a major role in the formation of the conducting layer. There appears to be some type of universality in the values of the 2D carrier concentration and of the thickness of the metallic sheet. The electronic states in the 2DEG can be endowed with physical properties that are observed in the bulk material, such as large spin-orbit, ferroelectricity, magnetic polarization, but in a surface specific way which reflects the orbital reconstruction and the spatial dichotomy of the energy bands. We will review the main experimental facts revealed by Angular Resolved Photoemission measurements and theoretical results of calculations based on ab initio techniques and tight-binding modeling. From the latter we will showcase the predicted existence of a 1D Z₂ spin-Hall topological states in the SrTiO₃ 2DEG for the (001) orientation.

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Atomic structure of the interface of superconducting monolayer FeSe on SrTiO₃

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The discovery of enhanced superconductivity in monolayer FeSe grown on SrTiO₃ by molecular beam epitaxy is an active area of research. A complete understanding of the atomic structure at the interface between FeSe and SrTiO₃ is critical to revealing the origins of the strong enhancement in T_c of this system. Here we report on *in situ* synchrotron x-ray diffraction experiments performed on superconducting FeSe/SrTiO₃. Crystal truncation rod (CTR) analysis reveals a $\sqrt{13} \times \sqrt{13}$ -R33.7° reconstruction at the SrTiO₃ surface with a double-TiO₂ layer termination. We extract the registry of the FeSe lattice with the SrTiO₃ substrate using a combination of CTR analysis and transmission electron microscopy. The structural parameters extracted from this analysis are important factors in guiding theories of superconductivity in monolayer FeSe.

X-ray structure analysis software for perovskite-type oxide thin films

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Structural information of the interfaces between perovskite oxides is vitally important to understand and control the functionality of oxide devices. For better control of interfacial structures, quick and user-friendly analyzing techniques for heterostructures are demanded.

Crystal truncation rod (CTR) scattering method, which is one of the surface X-ray diffraction methods, can provide the depth profile of the structure around the surface with sub-Å resolution. Although there are various ways to solve the phase problem have been proposed [1], the large number of structural parameters hampers us from obtaining a conventional method to provide the interfacial structure.

We have developed a convenient software to obtain the atomic arrangement of perovskite-type oxide thin films based on the reverse Monte-Carlo (RMC) method. The software can provide the wide acceptance of the initial model to converge to the global minimum of the χ^2 . The probability density of the interfacial structure, which contains atomic position and occupancy for each site, is estimated by the Bayesian inference. This method automatically provides the standard deviation of obtained structural parameters.

The figure shows a reported CTR profile of the 5-unit cell-thick LaAlO_3 ultrathin film made on SrTiO_3 substrate [2]. Calculated intensity profiles for the initial model and refined one by RMC method are also shown by the dashed and solid curves. Obtained real space structure reproduces the reported one [2]. The standard deviations of atomic positions were obtained to be $\pm 0.02\text{\AA}$ for metal sites and $\pm 0.03\text{\AA}$ for oxygen sites. The software has been applied to ten other datasets, including different substrate or film substances. It always provides a stable answer for the heavy elements, while the resulting oxygen positions are often unstable.

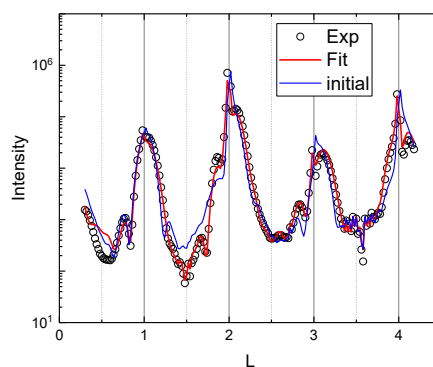


Figure. A CTR intensity profile of LaAlO_3 ultrathin film made on SrTiO_3 . Experimental results [2] are presented by the open circles, and the intensity profiles for initial and resulting models are presented by the dashed and the solid curves.

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Microscopic band structure of functional oxides

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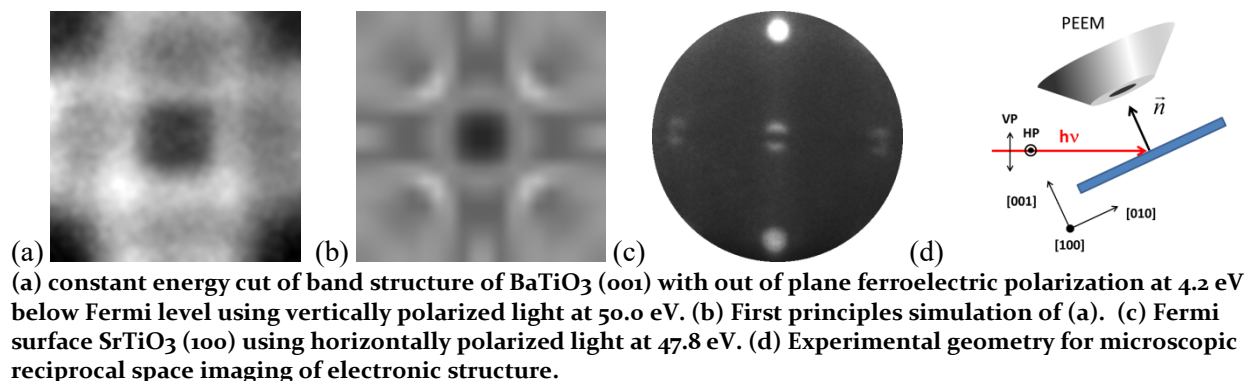
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Functional ferroelectric oxides and phenomena such as 2D electron gas (2DEG) at oxide interfaces represent potential technological breakthroughs for post-CMOS electronics.



Non-invasive techniques are required to study the surface chemistry and electronic structure underlying their often unique electrical properties. The sensitivity of photoemission electron microscopy (PEEM) to local potential, chemistry and electronic structure makes it an invaluable tool for probing the near surface region of microscopic regions and domains of functional materials

Following an introduction to PEEM we will illustrate two examples of analysis of the electronic band structure from microscopic regions of functional oxides.

In the first example, we present a spatial and wave-vector resolved study of the electronic structure of micron sized ferroelectric domains at the surface of a BaTiO₃ (001) single crystal. Reciprocal space imaging of the electronic structure using linearly polarized light provides unambiguous evidence for the presence of both in and out of plane polarization with two and fourfold symmetry, respectively. The results agree well with first principles calculations.

In the second example, we present results demonstrating a room temperature 2DEG at the surface of SrTiO₃ (100). The 2DEG is switched on by the soft X-ray irradiation and can be tuned by adjusting the redox conditions. At the same time reduction/oxidation of surface Ti is observed as a function of the oxygen partial pressure under beam.

This work has been supported by the Triangle de la Physique ELOXIR and the CEA Operando projects. We acknowledge Sincrotrone Elettra for the provision of synchrotron radiation facilities.

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Electronic transport in nanowires confined at the LaAlO₃/SrTiO₃ interface

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At the interface between LaAlO₃ and SrTiO₃ is confined a two dimensional electron system (2DES) characterized by several properties tunable by the electric field effect; among those, superconductivity, the 2DES undergoes a superconducting phase transition at ~300 mK, and spin-orbit coupling.

By using an atomic force microscopy (AFM) writing technique [1] the electrons at the interface can be confined in-plane; the conducting nanostructures defined by using this method have a lateral dimension as small as 50 nm [2].

The size of these devices is close to some characteristic lengthscales of the 2DES such as the electron mean free path and the superconducting coherence length. In this regime the interplay between the properties of complex oxides and the effect of dimensionality is increased and may lead to interesting physical effects.

The work presented here focuses on the comparison between the electronic transport in standard 2DES and in conducting nanowires realised by AFM-writing at the interface. The reduced dimensionality mainly affects the transport below ~ 30K where the behavior is strongly determined by the quantum corrections. Strong effects are also expected in the superconducting regime.

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Anisotropic transport and anomalous Hall effect in epitaxial spinel NiCo_2O_4 films

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The spinel complex oxides AB_2O_4 possess two distinct cation chemical environments, with the A-site surrounded by oxygen tetrahedral and the B-site accompanied by oxygen octahedral coordinates. This complex electronic structure has led to various interesting electronic, magnetic, optical and catalytic properties, which have been widely studied in the electrochemical area. As a prototype spinel oxide, the ferrimagnetic NiCo_2O_4 (NCO) has an inverse spinel structure with the Neel temperature of 673 K. It has been extensively studied as transparent electrodes and electrocatalysis due to its superior conduction characteristic¹.

Despite the active research carried out on NCO, the majority of the work has focused on various nanoparticles and nanostructures prepared by the chemical reaction techniques, while there are only few studies of epitaxial NCO thin films. In this work, we reported the epitaxial growth of high quality ultrathin NCO films and the observation of their unusual transport properties. The as-grown NCO films exhibit a highly anisotropic in-plane conductivity with an electrical dead layer of 2 u.c. (~1.6 nm), and possess an out-of-plane net magnetization. Anomalous Hall effect (AHE) has been observed at the measured temperature range from 2 K to 350 K, which changes sign at ~190 K. Measurements of the planar Hall effect reveal a uniaxial magnetic anisotropy that is not along any high symmetry crystal orientation. Our study reveals the complex energy landscape in NCO due to the competition of the crystalline field with the charge and spin degrees of freedom.

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Novel Two-Dimensional Silica Ferroelectric

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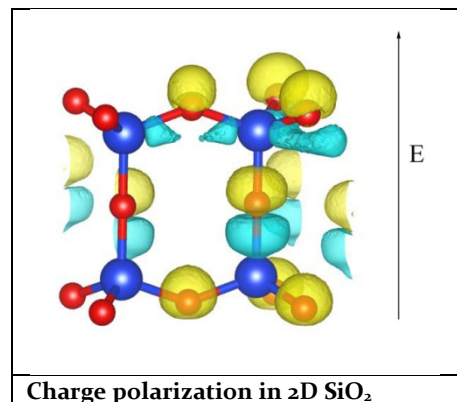
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Two-dimensional silicon oxide is a very important and promising material for nanoelectronics and optoelectronics [1]. However, the effect an external electrostatic field have on the polarization has not been studied. Therefore, in this study, we investigated in detail the atomic and electronic structure of 2D SiO₂ with different configuration, and compared the results with the available experimental data of the electronic and structural properties.

The first-principles calculations were performed with the generalised gradient approximation and spin polarization using density functional theory with the Perdew–Burke–Ernzerhof exchange-correlation functional in the Quantum Espresso software package.

We believe that our results of the structural and electronic properties are very important for researchers and technologists in the production and prospective use of nanostructured materials based on two-dimensional silicon oxide.



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Characterization of polar substrate surfaces using lateral force microscopy

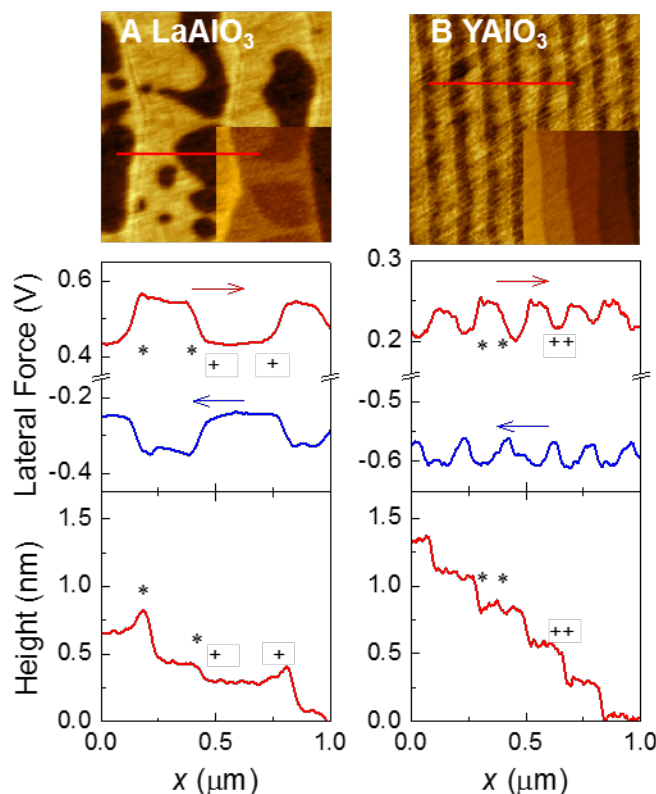
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Precise control of oxide substrate surfaces is key to generating advanced oxide nanodevices and thin-films. For example, the TiO_2 -terminated SrTiO_3 (STO) substrate surface is integral to the formation of the conducting 2DEG at the $\text{LaAlO}_3/\text{SrTiO}_3$ heterointerface.¹ So far, single-terminated oxide substrates have dominated oxide thin-film research. However, for polar perovskite oxides where mixed terminations can form after high-temperature annealing², selective thin-film deposition may be realized. Typically, these surfaces are dual-terminated randomly, thus limiting potential nanodevice applications.³

We have investigated (110)-oriented LaAlO_3 (LAO) and (110)-oriented YAlO_3 (YAO) substrate surfaces using lateral force microscopy, as shown in panel A and B, after annealing at 1000 °C for 1 hr in air. Unlike LAO (A), YAO forms nanostripes between its two terminations (B). The LAO substrate shows clear half-unit cell steps between differing frictions whereas the YAO substrate does not, which is due to limitations in the AFM tip resolution³ and the substrate miscut angle. Thus YAO substrates, with their ordered dual-terminated nanostripes, are potential new candidates for selective thin-film deposition.



Lateral force microscopy of an LAO (A) and YAO (B) substrate after annealing. The inset shows the corresponding AFM signal. The lateral force and line profile are shown below each image with the red and blue arrows indicating the scan direction. * and + indicate areas of differing friction. Images are $1.5 \mu\text{m}^2$.

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Tunable metal-to-insulator transition in $\text{La}_{1-x}\text{Nd}_x\text{NiO}_3$ thin films

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To elucidate the role of the NiO_6 octahedral rotation on transport and magnetic properties, we have grown a series of coherently strained, 60 unit cell thick, $\text{La}_{1-x}\text{Nd}_x\text{NiO}_3$ single layer alloys (Fig. 1) on (001) SrTiO_3 . Growth on (001) SrTiO_3 by pulsed laser epitaxy imparts a nominal tensile strain ranging from 1.9-2.4% depending on x . A La-doped NdNiO_3 alloy will see a slight reduction in ϵ as pure LaNiO_3 has a tensile strain of $\epsilon(\text{bulk}) \approx 1.9\%$. Transport measurements using the Van der Pauw configuration indicated three distinct regions of resistivity: i) an insulating state with no metal-to-insulator transition ($x = 0$), ii) a measurable metal-to-insulator transition ($x = 1/8$ - $4/8$), and iii) metallic state with no metal-to-insulator transition ($x > 4/8$). As seen in figure 1, pure NdNiO_3 is an insulator at all temperatures, contrary to previous experiments of NdNiO_3 under tensile strain[1]. With the introduction of La (likely via site substitution), we observed a metal-to-insulator transition that decreased with decreasing x . In addition, a transition that appears to be first order with the width of the temperature hysteresis decreasing with increasing La content was also observed. Above $x = 4/8$, the thin films appear to be metallic down to the lowest temperatures measured, 2 K. Resonant elastic x-ray scattering experiments indicate that the films have the typical $\mathbf{q}=(1/4,1/4,1/4)$ antiferromagnetic ordering with Néel temperatures similar to the metal-to-insulator transition temperature, in agreement with previous experiments[2]. In this work, we have shown that with addition of La, an atom with a slightly larger ionic radius (compared to Nd), results in the gradual transition from the insulating phase to the metallic phase; the result of a systematic reduction in the tolerance factor leading to a change in the NiO_6 octahedra rotation.

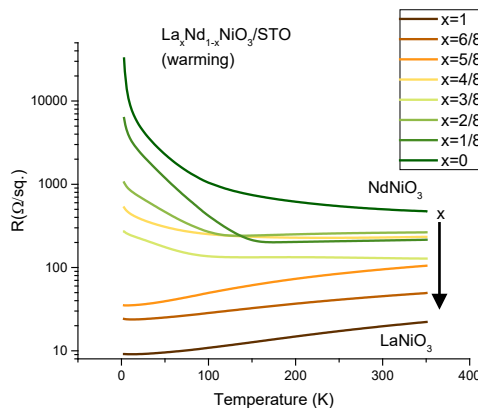


FIGURE 1: RESISTIVITY AS A FUNCTION OF TEMPERATURE FOR $\text{La}_x\text{Nd}_{1-x}\text{NiO}_3$ FILMS ON SrTiO_3 (001) SUBSTRATES. THE CURVES PLOTTED WERE OBTAINED UPON WARMING FROM 2 K AFTER COOLING IN ZERO APPLIED FIELD FROM 350 K.

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Correlation physics in high mobility MgZnO/ZnO heterostructures

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The MgZnO/ZnO heterointerface hosts a two dimensional electron system which display electron mobilities in excess of 1,000,000 cm²/Vs. This has been realized due to improvements in sample growth techniques over the past decade [1,2]. Figure 1 plots these advances with the mobility (μ) plotted as a function of charge density (n). The peak in μ for each generation shifts higher as the charged impurity content is reduced and the interface is made cleaner.

In this presentation we canvas the state of the art growth techniques, along with the correlated ground states that the high mobility electrons elicit at ultra-low temperatures. The two-dimensionality of carriers enables the observation of the quantum Hall effect with distinct features of the strong correlation effects of oxides.

One outcome is parameter renormalization of the Fermi-liquid. This manifests as an enhancement of the effective mass as n is reduced, which acts to suppress the cyclotron energy splitting of electrons. By tuning the Zeeman energy, we are able to selectively control transitions between the discrete orbitals and spin orientation of carriers. This has spectacular results in the fractional quantum Hall regime, where we manage to induce new ground states which take an even-denominator filling at $\nu = 3/2$, which may host anyons who obey non-Abelian statistics [3]. Further, while similar tunability is observed at $\nu = 5/2$, we find that nematic phases which break rotational symmetry are in close competition. These “stripes” appear as anisotropic magnetotransport and display strong magnetic field sweep direction hysteresis, potentially indicating an Ising ferromagnet-like spin texture [4].

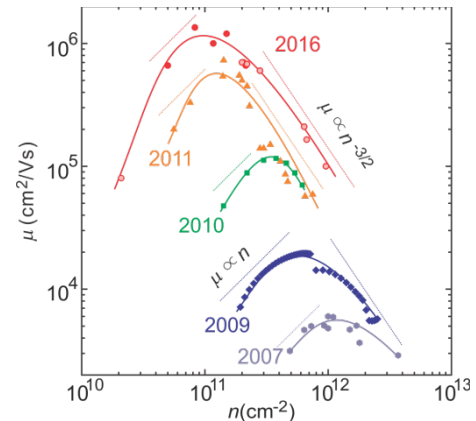


Figure 1: Mobility (μ) of ZnO-based two dimensional electrons as function of charge density (n) for multiple generations of growth technology. The guide-lines of $\mu \propto n$ and $\mu \propto n^{-3/2}$ correspond to impurity and interface scattering mechanisms, respectively. Here, $T \leq 2$ K.

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Exploring the phase diagram of $\text{La}_{1-x}\text{Nd}_x\text{NiO}_3$ thin films

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In this work we present a series of results on thin films of $\text{La}_{1-x}\text{Nd}_x\text{NiO}_3$. The phase diagram spanned by this mixed nickelate compound extends from metallic, paramagnetic and rhombohedral LaNiO_3 for $x = 0$ to NdNiO_3 , an orthorhombic perovskite with a metal-charge transfer insulator transition for $x = 1$ [1].

The good quality materials are grown by radio frequency off-axis magnetron sputtering co-deposition, using two stoichiometric ceramic targets of the end materials, constituting an important materials growth study.

Structural information is obtained by x-ray diffraction and the electronic properties are accessed by low-temperature transport. The evolution of the physical properties is then traced across the phase diagram in an effort to understand the onset of the metal-insulator transition for which the nickelate family has become so famous [2].

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Anisotropic electronic transport of the two-dimensional electron system in $\text{Al}_2\text{O}_3/\text{SrTiO}_3$ heterostructures

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Transport measurements on the two-dimensional electron system in $\text{Al}_2\text{O}_3/\text{SrTiO}_3$ heterostructures indicate significant non-crystalline anisotropic behavior below $T \approx 30$ K. Lattice dislocations in SrTiO_3 and interfacial steps are suggested to be the main sources for electronic anisotropy. Anisotropic defect scattering likewise alters magnetoresistance at low temperature remarkably and influences spin-orbit coupling significantly by the Elliot-Yafet mechanism of spin relaxation resulting in anisotropic weak localization. Applying a magnetic field parallel to the interface results in an additional field-induced anisotropy of the conductance, which can be attributed to Rashba spin-orbit interaction. Compared to $\text{LaAlO}_3/\text{SrTiO}_3$, Rashba coupling seems to be reduced indicating a weaker polarity in $\text{Al}_2\text{O}_3/\text{SrTiO}_3$ heterostructures.

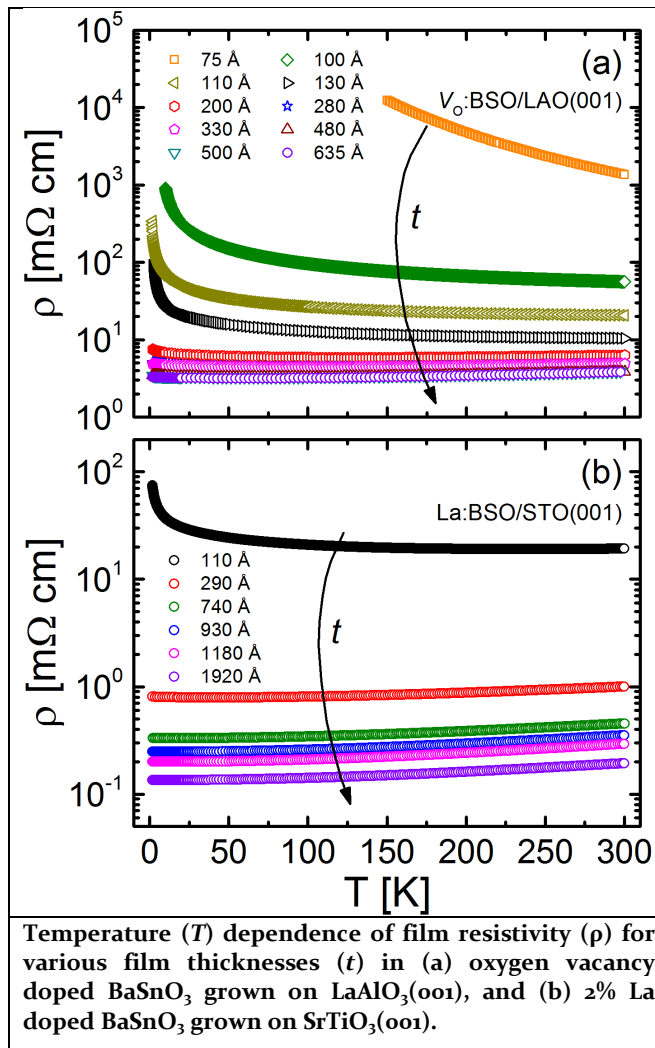
Mobility Optimization and Electronic Transport in High Pressure Oxygen Sputter-Deposited BaSnO₃ Thin Films

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The recent discovery of high room temperature mobility in wide band gap BaSnO₃ has generated exceptional interest in this perovskite oxide for electronic devices. Outstanding issues with regards to epitaxial films include understanding transport behavior, determining the optimal dopant, and understanding the role of structural defects (like dislocations) in limiting mobility. Here, we present detailed temperature and field-dependent electronic transport in both oxygen vacancy and La-doped BaSnO₃ (BSO) films grown via high pressure oxygen sputter deposition. High-resolution X-ray diffraction, scanning probe microscopy, and scanning transmission electron microscopy confirm phase-pure, close to stoichiometric, smooth, epitaxial BaSnO₃(001). Film thickness, growth rate, deposition temperature, and substrate (*i.e.*, lattice mismatch) have all been systematically varied, and related to mobility. [1] As-grown undoped, insulating films can be made conductive with *controllable* *n*-type doping by vacuum reduction, resulting in 300 K Hall mobilities up to 35 cm²V⁻¹s⁻¹ (on MgO) at 5×10¹⁹ cm⁻³. [1, 2] The mobility-electron density relation has been probed in this manner, down to 2×10¹⁷ cm⁻³. 2% La-doped BaSnO₃ films, on the other hand, demonstrate 300 K electron mobilities up to 60 cm²V⁻¹s⁻¹ at 3×10²⁰ electrons per cm³. With increasing film thickness a clear insulator-metal transition is observed with both dopants (see figure), likely related to defect density near the substrate. The results provide insight into the structure-electronic property relationships, mobility-electron density relation, and mobility-limiting factors in BaSnO₃ films.



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Strain Effects on Ionic Transport in Perovskite Oxides

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Engineering perovskite ABO_3 oxides to achieve superior oxygen ion conductivity has attracted considerable attention for applications in intermediate-temperature solid-oxide electrolysis cells. Epitaxial strain has long been suggested and predicted as a promising strategy to enhance oxygen ion diffusivity in perovskite oxides. Few systematic experimental studies on coherently-strained versions of these materials, however, have been conducted. At the same time, there has been limited understanding of the detailed diffusion mechanism(s) in such strongly correlated systems, and especially the correlation between strain-induced structural changes in perovskites (*i.e.*, unit cell volume, lattice distortion, and octahedral rotations) and ionic conductivity has yet been established theoretically or experimentally.

In this work, we focus on a model perovskite oxygen ion conducting system $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.9}\text{Mg}_{0.1}\text{O}_{3-\delta}$ (LSGM) and perform a systematic study of strain effects on ionic transport. Epitaxial strain (ranging from compressive -1.1% to tensile +0.79%) is applied by growing 100 nm LSGM thin films on various perovskite substrates (NdGaO_3 , LSAT, SrTiO_3 , DyScO_3) with pulsed-laser deposition. X-ray diffraction studies reveal single-phase, coherently-strained LSGM thin films under all strain conditions and Rutherford backscattering spectrometry studies confirm the film chemistry to be $\text{La}_{0.9}\text{Sr}_{0.1}\text{Ga}_{0.95}\text{Mg}_{0.05}\text{O}_{3-\delta}$. Structural characterizations with both laboratory and synchrotron X-ray sources reveal that the LSGM unit cell volume (lattice monoclinic tilt) monotonically increases (decreases) upon transitioning from compressive to tensile strain, while the octahedral rotation is strongest at zero strain but is largely quenched at both compressive and tensile strains. To correlate these strain-induced structural changes with ionic transport, electrochemical studies were performed with an impedance analyzer on films with Pt interdigitated electrodes. The ionic migration barriers for the heterostructures under different strain states were determined to be 0.85 eV (-1.10%), 0.73 eV (-1.05%), 0.65 eV (+0.1%), and 0.76 eV (+0.79%), respectively. Our results reveal that, strain can impose a dramatic change (~30%) on ionic migration barriers in perovskite materials. Such changes cannot be simply explained by the volumetric change of the unit cells, but are likely correlated to both lattice distortion and octahedral rotation effects. Importantly, it is found that larger unit cell volumes and stronger octahedral rotations are favorable for high ionic conduction. Our findings provide novel insights into the effects of structural motifs on ionic transport properties in perovskite materials, and shed light on the design guidelines for fast ionic conducting materials.

Engineering magnetism and polarization domain structures in BiFeO_3 containing thin films through superlattice structuring

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We report the growth of $\text{BiFeO}_3/\text{La}_x\text{Bi}_{1-x}\text{FeO}_3$ superlattices as well as the results of neutron diffraction and reflectometry measurements of the magnetic structure of these films. We have synthesized superlattice films using reactive oxygen molecular beam epitaxy (MBE) with a variety of La-doping concentrations in the La-doped BiFeO_3 layers. We describe these films as $y*[n*\text{BiFeO}_3/m*\text{La}_x\text{Bi}_{1-x}\text{FeO}_3]/\text{substrate}$ where n is the number of unit cells of BiFeO_3 , m is the number of unit cells of $\text{La}_x\text{Bi}_{1-x}\text{FeO}_3$, x is the La doping concentration, y is the number of superlattice repeats, and our substrate is either $\text{Nb:SrTiO}_3(100)$, $\text{SrTiO}_3(100)$, or $\text{TbScO}_3(110)$. X-ray diffraction and TEM measurements illustrate the high crystal quality of these films with this growth technique, and we have used high resolution STEM to image the local atomic displacements representative of the local unit cell polarizations, as shown in figure 1. We report the tuning of local displacements with changing superlattice parameters. We have also performed both neutron diffraction and reflectometry measurements at the NCNR at NIST.

Our neutron diffraction measurements show an unusual magnetic field dependence of the expected G-type antiferromagnetic structure for certain combinations of n , m , x , y , and substrate choice. Polarized neutron reflectometry allows us to probe the magnetic and nuclear depth profile of these superlattices. Using this technique combined with fitting to various magnetic depth profile models, we observe a non-zero magnetization within certain superlattices despite apparent G-type antiferromagnetic ordering from diffraction.

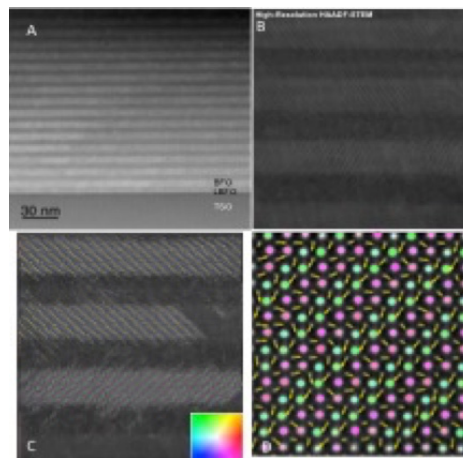


Figure 1; TEM measurements for an $n=m=14$ $\text{BiFeO}_3/\text{La}_x\text{Bi}_{1-x}\text{FeO}_3$ superlattice. a,b: HAADF-STEM images of the superlattice c: Local a-site distortion mapped onto the color square in the bottom right corner. The color shows the direction of displacement d. Color map with added local polarization vectors mapped.

Low-Temperature Dielectric Anisotropy Driven by an Antiferroelectric Mode in SrTiO_3

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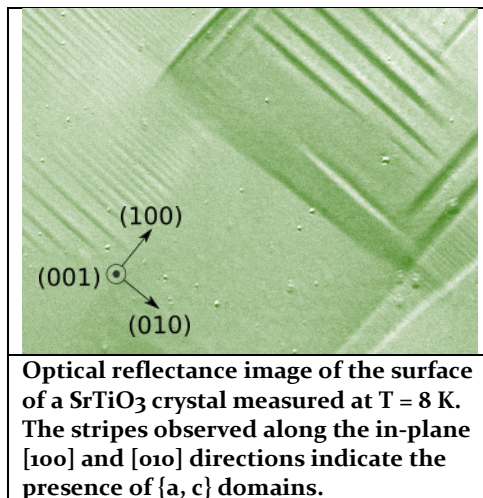
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Strontium titanate (SrTiO_3) is the quintessential material for oxide electronics. One of its hallmark features is the transition from cubic to tetragonal symmetry, below which antiferrodistortive (AFD) instabilities dominate and ferroelastic twins appear. Below this structural transition two types of ferroelastic twins can emerge on the surface of a (001)-oriented SrTiO_3 crystal. On the one hand, {a} and {c} domains may arise in which the elongated axes are in-plane and out-of-plane, respectively. Alternatively, {a₁} and {a₂} domains denote {a} domains having the elongated axis along orthogonal directions within the plane. We have used optical imaging to reveal the emergence of these twins (see Figure) and study their spatial distribution under in-situ applied electric fields ¹. We find that the field-induced motion of ferroelastic twins is governed by an anisotropic dielectric constant where the softer component is normal to the tilt axis of the AFD mode. From first-principles and Landau theory we find that this anisotropy can be explained unambiguously by the emergence of a tri-linear coupling involving antiferroelectric (AFE), polar and AFD lattice distortions ¹. Interestingly, the appearance of this trilinear coupling also solves the longstanding issue of the origin of the R-point infrared (IR)-active phonon at $\sim 440 \text{ cm}^{-1}$, which is commonly used to infer the formation of ferroelastic twins in SrTiO_3 crystals ². So far, this R-point phonon mode was generically assigned to the back-folding of the Brillouin zone in the tetragonal phase. Our study clarifies definitely the origin of this mode and gives an accurate representation for the lattice deformations that give rise to its emergence.



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Flexoelectricity induced large tunable selfbias by a PZT interfacial layer of epitaxial PMN-PT ferroelectric capacitors

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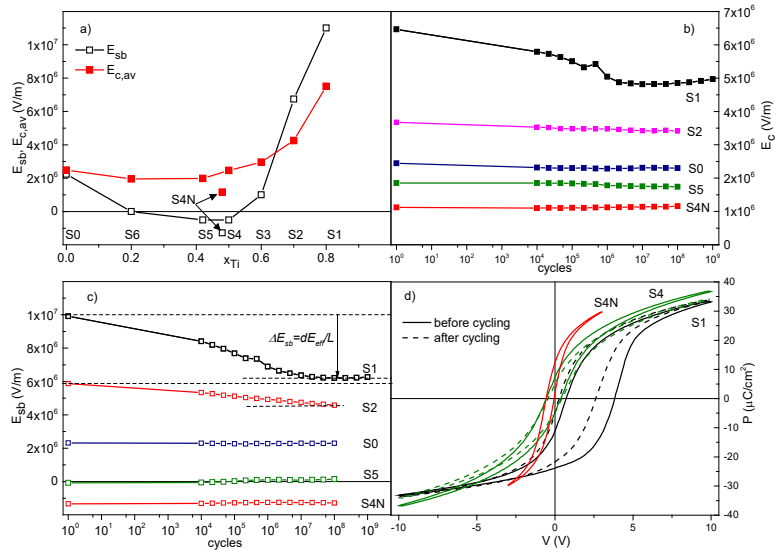
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Epitaxial $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$ (PMN-PT) 200 nm thick, (001) oriented, phase-pure films were grown on a range of $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ (PZT) buffer layers ($x=0.2\text{-}0.8$) and directly on the SrRuO_3 electrode on (001) SrTiO_3 substrates in a ferroelectric capacitor structure.

For some of the PZT compositions extremely high strain is obtained in the 25 nm thick buffer layer, with c/a ratio up to 1.09, as was deduced from XRD experiments. This strain partly relaxes over the thickness of this layer, creating a strain gradient.

Strain gradients can cause the flexoelectric effect, the induction of a permanent polarization vector. The PMN-PT devices with a highly tetragonal PZT buffer layer and without buffer layer show very large self-bias fields up to 1.0×10^7 V/m. These self-bias fields correlate with the strain gradient layers near the bottom electrode observed in these devices only. We interpret the large self-bias quantitatively in terms of the flexoelectric effect. An alternative explanation may be the presence of trapped charges in the strain gradient layer.

Fatigue measurements show that the self-bias field decreases with increasing number of cycling of the ferroelectric hysteresis loop. This can be explained by compensation of the self-bias field arising from the permanent polarization by permanent, slowly diffusing charges that are accumulating at the top of the buffer layer.



(a) self-bias field E_{sb} and coercive field E_c (before aging) of PZT/PMN-PT capacitors. Lines are guides to the eye. (b) E_c and (c) E_{sb} versus number of cycling (up to 5×10^7 V/m); (d) polarization loops before and after cycling.

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Search for superconductivity in the infinite-layer CaCuO_2 system

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Infinite-layer CaCuO_2 (CCO) is the essential structure of high temperature superconductors, e.g., Bi-2212. In those CCO layers, Cu ions are square-planar coordinated. Moreover, those CCO layers, are sandwiched by SrCuO_2 where Cu ion is octahedral coordinated. Disentangling the correlations of the two intertwined Cu interactions is of foremost importance in order to derive models that have potential not only to describe current but also to predict future superconductors. This disentanglement requires the preparation of CCO in an isolated form. However, the synthesis of single crystals of CCO itself is a major challenge in materials science and we therefore employed molecular beam epitaxy (MBE), which allowed us the growth of single-crystalline CCO films. Here, we show that for randomly Sr substituted CCO films, superconductivity is induced by an annealing procedure under oxidizing condition but is limited to the surface region [1]. This leaves us with two possible scenarios on how the induction of superconductivity takes place in the Sr substituted CCO films. First, there are cooperative correlations between square planar and octahedral coordinated Cu ions where the layer containing the octahedral coordinated Cu ions act as a charge reservoir. Namely, Cu ions in the topmost layer are octahedral coordinated and the subsequent layer, containing square-planar coordinated Cu, is superconducting. On the other hand, it is possible that the annealing procedure necessary for the induction of superconductivity eradicates oxygen vacancies only in the surface region. Taking also into account reports by Castro *et al.* [2], the first scenario seems more plausible. We also show that carrier doping into CCO films by a partial substitution of Nd for Ca results in a surge in electron conductivity by at least four orders of magnitude. Furthermore, a reduced MBE growth rate additionally boasts the electronic conduction triggered by a reduction of oxygen vacancies. The synthesis of superconducting CCO single crystals, while still elusive, would pave the way for exploring new superconductors by mimicking the crystal structure of high temperature superconductors.

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Charge confinement in epitaxially grown $\text{LaTiO}_3/\text{SrTiO}_3$ films on Si by multi-band Hall measurements

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The integration of complex oxides onto semiconductors has opened up an attractive field of research, combining the multifunctionality of oxide systems with well-established semiconductor platforms. One limitation of electronics based on oxide systems is typically a phonon-limited carrier mobility at room temperature. We demonstrate an approach to remotely dope into silicon from a 2-dimensional electron gas (2DEG) formed at the interface between LaTiO_3 and SrTiO_3 . By introducing oxygen at the interface between SrTiO_3 and Si, we show that we can raise the conduction band offset by ~ 0.5 eV. We also study the effect of changing the 2DEG confinement by varying the SrTiO_3 layer thickness by fitting transport data to a multi-band Hall model, showing that the SrTiO_3 forms a well that traps electrons, pulling charge away from the silicon. With this understanding we can better design a materials system to control the movement of charge carriers in order to achieve a high mobility oxide-semiconductor system.

Peculiar oxygen and copper isotope effects on the pseudogap formation temperature in underdoped to overdoped cuprates: Pseudogap induced by pairing correlations above T_c in cuprates with large and small Fermi surfaces

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We investigate the pseudogap (PG) state and the peculiar oxygen and copper isotope effects on the PG onset temperature T^* in cuprate superconductors with large and small Fermi surfaces within the polaron model and two different BCS -based approaches extended to the intermediate coupling regime. We argue that the unconventional electron-phonon interactions are responsible for the polaron formation and BCS -like pairing correlations above T_c in underdoped to overdoped cuprates, which are exotic (non- BCS) superconductors. Using the generalized BCS -like theory, we calculate pseudogap formation temperatures T^* , isotope shifts ΔT^* , oxygen and copper isotope exponents (i.e. $\alpha_{T^*}^O$ and $\alpha_{T^*}^{Cu}$) and show that isotope effects on T^* strongly depend on strengths of Coulomb and electron-phonon interactions, doping levels and dielectric constants of the cuprates. This theory explains the existence of small positive or sign reversed oxygen isotope effect, sizable and very large negative oxygen and copper isotope effects on T^* in cuprates with large Fermi surfaces. Further, we use another version of the extended BCS -like model to study the PG formation and the peculiar isotope effects on T^* in deeply underdoped cuprates with small Fermi surfaces and predict the existence of small and sizable negative oxygen and copper isotope effects on T^* in such underdoped cuprates. The results for T^* , isotope shifts ΔT^* and exponents ($\alpha_{T^*}^O$ and $\alpha_{T^*}^{Cu}$) in different classes of high- T_c cuprates are in good agreement with the existing well-established experimental data and explain the controversy between various experiments on isotope effects for T^* in the cuprates.

Atomic Structure Evolution of $\text{Sm}_{1-x}\text{Sr}_x\text{TiO}_3$ at Mott transition

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Perovskite rare-earth titanates (RTiO_3 , where R is a trivalent rare earth element) are of significant interest for understanding emergent phenomena caused by strong electron correlations. They are prototypical Mott insulators. Their properties are determined by the degree of the orthorhombic distortion in the perovskite crystal structure. For example, it determines the critical doping density required to induce the metal-to-insulator transition (MIT). However, despite the importance of the correlation between structure and material properties, a lack of experimental studies exists regarding the microscopic understanding of mechanisms for physical phenomena observed in RTiO_3 -based Mott insulators. Using scanning transmission electron microscopy (STEM), we study the doping dependent structural change in $\text{Sm}_{1-x}\text{Sr}_x\text{TiO}_3$ films grown on LSAT substrates, in which the MIT occurs with the Sr content of $0.05 < x < 0.10$.

SmTiO_3 is a Mott insulator with a distorted perovskite structure (space group $Pbnm$). Imaged along $[110]_0$, where the subscript indicates the orthorhombic unit cell, Sm displacements in a “zig-zag” pattern are seen as shown in Fig. 1 (a). Using this, we measure the deviation angle ($180^\circ - \theta_x$) formed by three successive Sm atomic columns to quantify structural distortions. Shown in Fig. 1 (b) are distributions of deviation angles from $\text{Sm}_{1-x}\text{Sr}_x\text{TiO}_3$ films with $x = 0, 0.05$, and 0.10 , among which only $\text{Sm}_{0.90}\text{Sr}_{0.10}\text{TiO}_3$ film shows metallic behavior. As the Sr content increases, the deviation angles gradually decrease, indicating reduced structural distortions due to Sr doping. Here, the standard deviations of deviation angles represent the measurement error (e.g. scan distortion and sample drift during STEM imaging) and any inhomogeneity within the film (e.g. phase separation or defects). The standard deviation values of Sr-doped SmTiO_3 films are comparable to that of the stoichiometric SmTiO_3 film and all three samples adopt an orthorhombic distorted perovskite structure. Thus, our measurements on atomic column positions provide direct evidence of continuous and homogeneous structural changes with the absence of phase separation. This finding presents detailed structural information with high spatial resolution and precision and provides an important step toward a complete understanding of unconventional physical phenomena near Mott transitions.

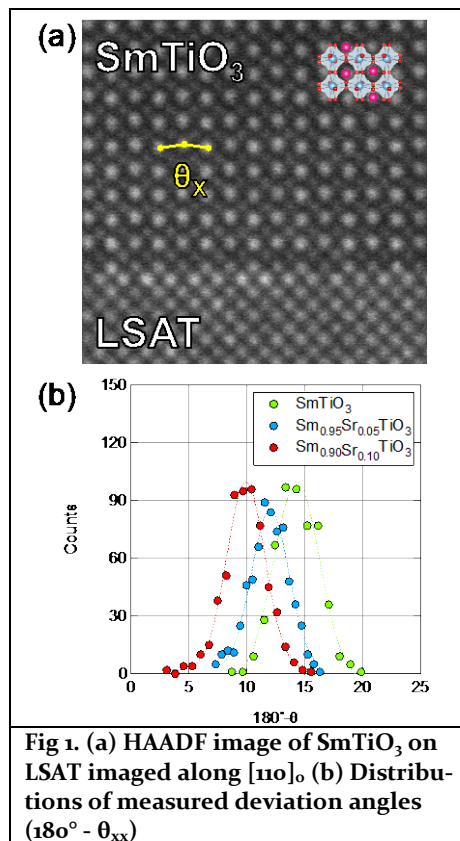


Fig 1. (a) HAADF image of SmTiO_3 on LSAT imaged along $[110]_0$ (b) Distributions of measured deviation angles ($180^\circ - \theta_{xx}$)

Epitaxial ferroelectric varactors for low-voltage microwave applications

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In metal-insulator-metal (MIM) parallel-plate ferroelectric varactors, the permittivity of the functional oxide dielectric $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$ ($0 < x < 1$, BST) layer can be tuned by an electric field applied between the conducting electrodes. In this work, we present a technology of the Au/Pt/BST/SrMoO₃ varactors for all-oxide low-voltage microwave tunable devices based on SrMoO₃ as novel electrode material. This material has two electrons in a wide 4d (t_{2g}) band near the Fermi level and shows a low room-temperature resistivity of 5 $\mu\Omega\cdot\text{cm}$. The BST/SrMoO₃ varactor heterostructures were epitaxially grown on GdScO₃ substrates using pulsed laser deposition controlled by reflective high-energy electron diffraction (RHEED). The observed RHEED intensity growth oscillations reveal the 2D growth mode of the varactor heterostructures with remarkably thick SrMoO₃ films of up to 2.7 μm and 40-300 nm thick BST films. The perfection of the crystal structure was confirmed by the high-order Laue fringes observed in the x-ray diffraction patterns of the produced thin-film samples. The STEM/EELS studies show sharp interfaces in the heterostructures with intermixing of the materials at the distance of one unit cell. As the last fabrication step, the 300/20nm thick Au/Pt thin-film top electrodes were deposited and patterned using standard photolithography lift-off process.

The electric tunability $n=C(0)/C(3.7\text{V}) \approx 4$ of the varactors is achieved at battery-low voltages due to the used thin BST functional layers. Such high tunability indicates that the effective BST stoichiometry control can be achieved with fine tuning of the growth parameters. Low losses of the varactors at gigahertz frequencies originate from the used thick SrMoO₃ bottom electrodes with resistivity of about 30 $\mu\Omega\text{cm}$ and result in the quality factor of the varactors Q higher than 30 at 1 GHz. The losses due to the GdScO₃ substrates are rather small as the SrMoO₃ thickness is comparable to the skin depth of SrMoO₃ at gigahertz frequencies. The developed Au/Pt/BST/SrMoO₃ epitaxial oxide varactors open a path for realization of all-oxide microwave devices and components.

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Structural characterization of the $\text{LaInO}_3/\text{BaSnO}_3$ interface via synchrotron scattering

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The alkaline earth stannate BaSnO_3 has a large room-temperature mobility for a conducting perovskite oxide. Field effect transistors (or FETs) based on BaSnO_3 display a large on/off ratio, $I_{\text{on}}/I_{\text{off}}=10^7$, and high field effect mobility, $\mu \sim 90 \text{ cm}^2/\text{Vs}$. These devices use a polar LaInO_3 dielectric in which it has been suggested that the polar discontinuity between the orthorhombic and polar LaInO_3 dielectric and the cubic and nonpolar $\text{Ba}_{0.93}\text{La}_{0.07}\text{SnO}_3$ channel leads to an electronic reconstruction. LaInO_3 remotely dopes $\text{Ba}_{0.93}\text{La}_{0.07}\text{SnO}_3$ with electrons, creating the carriers for these high mobility devices.

We determine the role of rotations of oxygen octahedra in influencing the polarization of LaInO_3 using synchrotron x-ray diffraction. These rotations are characterized by measuring half-order Bragg peaks of thin film $\text{LaInO}_3/\text{BaSnO}_3/\text{SrTiO}_3$ heterostructures grown by pulsed-laser deposition. We verify that these measured peaks are consistent with the octahedral rotation pattern of bulk LaInO_3 , which has Glazer tilt pattern of $a^+b^-b^-$. Fitting these peaks, we determine the rotational angles as a function of LaInO_3 film thickness and investigate the effect of the rotation on the $\text{LaInO}_3/\text{BaSnO}_3$ interface. We discuss how these rotations may be coupled to interfacial polarization and the observed transport properties.

Observation of metallic-like ferroelectricity in ~1 nm thick deficient BaTiO₃ at room temperature

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Requirements of multi-functionalities in thin-film systems have led important findings of unique physical character and degree of freedom which only exist in film forms. As growth technique gets advanced, one can decrease the film thickness even several nano-meter (~nm) scale where its unique physical character still appears. Among those intriguing film systems, ferroelectric has been of interest. As a prototype ferroelectric, electrical properties of ultra-thin BaTiO₃ (BTO) films have extensively studied, which is found that ferroelectricity sustains down to ~ nm-thick films as theoretically predicted. However, efforts on determination of the minimum thickness in ferroelectric films has been hindered by large leakage current. In this study, we used ~ nm-thick BTO films showing metallic-like behaviour around room temperature (RT). We found that even the 2 unit-cells-thick metallic-like BTO film shows ferroelectric switching behaviour at RT. Observation of such ultra-thin conducting ferroelectric will enlarge its applicable fields leading realization of new functional devices and investigations of further physical phenomena.

Strain control of oxygen kinetics in epitaxial Ruddlesden-Popper oxides

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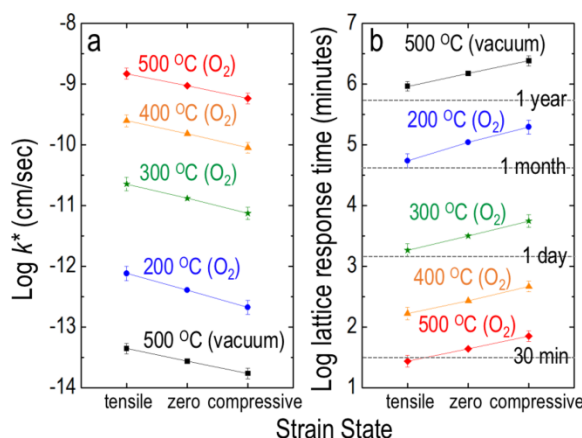
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Oxygen defect control has long been considered an influential tuning knob for producing various property responses in complex oxide films. However, the nature of oxygen defects in thin films is often not investigated beyond basic redox chemistry. One of the model examples for oxygen defect studies is the layered Ruddlesden-Popper (RP) phase $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-\delta}$ (LSCO), in which the superconducting transition temperature (T_c) of epitaxial thin films was previously found to be highly sensitive to epitaxial strain. However, previous observations on strain-superconductivity coupling in LSCO thin films were mainly understood in terms of elastic contributions to mechanical buckling, with minimal consideration of kinetic or thermodynamic factors. Here, we report that the oxygen nonstoichiometry that is commonly reported for these strained cuprates is mediated by the strain-modified surface exchange kinetics, rather than reduced thermodynamic oxygen formation energies for one strain state versus another. Both of these are highly correlated with the variability of the superconducting transition in LSCO. Remarkably, tensile-strained LSCO shows nearly an order of magnitude faster oxygen exchange rate than a compressively strained film, revealing a strong contrast in the time scales required to modify oxygen stoichiometry. Since strain engineering is a very powerful tool for controlling oxygen surface exchange kinetics in Ruddlesden-Popper oxides, it is an enticing strategy for developing high-performance ionotronic materials and devices in addition to the controlling superconductivity.

This work was supported by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), Materials Sciences and Engineering Division (experiment) and by the NSF Software Infrastructure for Sustained Innovation (SI2) award No. 1148011 (theory).



Strain-dependent oxygen kinetics. Variation of predicted oxygen surface exchange rate k^* (a) and time required to incorporate oxygen (b) under different annealing conditions and strain states. Overall, k^* is larger in tension than in compression. Conversely, the shortest oxygen filling times correspond to tensile strain, and the longest times to compressive strain as shown in b.

Modification of Magnetocrystalline Anisotropy via Ion-Implantation

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Spin textures are defined by arrangements of magnetic moments within nano- or microscale patterned features within a magnetic material. Artificial skyrmions are particularly intriguing and Li *et al.* demonstrated they can be created by fabricating Co nano-pillars with in-plane moments that form a magnetic vortex state, surrounded by a Ni film with out-of-plane magnetization (1). An analogous effect can be achieved in a thin film of a single material if the magnetocrystalline anisotropy can be locally modified. Ion-implantation of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ thin films can induce changes in anisotropy by causing an out-of-plane uniaxial lattice expansion while the film remains coherently strained to the substrate, but reduces the Curie temperature and saturation magnetization of films. X-ray absorption spectroscopy shows this degradation in magnetic properties is in part due to the reduced oxidation state of Mn after implantation. By tuning implant and post-implant annealing conditions, as well as proper substrate selection, the easy axis of the film can be tuned between in and out-of-plane while minimizing the reduction in magnetism.

The research reported in the paper has in part been supported by the National Science Foundation (DMR 0747896 and 1411250).

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Dimensionality control of a novel electron gas based on KTaO_3 (001)

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The discovery of two-dimensional electron gases (2DEGs) at transition metal oxide (TMO) surfaces and interfaces has opened up broad interest due to their exotic properties such as quantum Hall effect, 2D superconductivity and gate controlled ground states. Recently, 5d TMOs are hotly investigated due to their strong spin-orbit coupling (SOC), a key element of topological materials. Among them, KTaO_3 (KTO) not only hosts 2DEGs but also involves strong SOC. Here we report the discovery of electron gas based on KTO oxide interface, with low temperature mobility as large as $8000\text{cm}^2\text{V}^{-1}\text{s}^{-1}$. Strong Shubnikov-de Haas (SdH) oscillation in magnetoresistance is observed at low temperatures. Based on this playground we demonstrate a novel technique to perform quantum confinement engineering by inserting an insulating spacing layer into the interface. Indeed, we observed a drastic change in SdH oscillation from 3D-like behavior to 2D-like behavior. In addition, Fermi surface reconstruction due to the quantum confinement is also observed from SdH oscillation. Our results not only provide a novel playground for condensed matter physics and all-oxide device applications, but also open a promising new route in tailoring the dimensionality of electron gas systems.

Optical visualization of oxygen vacancies migration in Ca doped bismuth ferrite

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Controlling the oxygen vacancies is important to construct expected physical properties. In resistive switching memory, concentration of oxygen vacancies transforms the electronic structure to make a conducting state and controlled by external electric field. In addition, solid oxide fuel cells directly convert stored chemical energy to usable electrical energy by transferring oxygen ions into electrolyte materials. As-grown Ca doped BiFeO₃ (BFO) is the one of the best oxide material to study motion of oxygen vacancies using an application of external electric field. Ca doped BFO spontaneously generates much of oxygen vacancies to stabilize an oxidation number of Fe³⁺ ions. Simultaneous measurement of time-resolved current value and x-ray diffraction notices topotactic transition induced by the migration of oxygen vacancies. X-ray absorption spectroscopy (XAS) and photoelectron emission microscopy (PEEM) experiments are carried out to observe their electronic structures in formed state. Furthermore, we trace the propagation of oxygen vacancies through the optical microscope and investigate the dynamics of oxygen vacancies along the temperature. Relationship between their dynamics and evolved physical properties help to design further application to control expected states. Besides, we suggest a novel way to define the physical value related to dynamics of oxygen vacancies through the optical microscope.

Emergent chirality in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices

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Recent work has shown the formation of three-dimensional polar vortices in ferroelectric/paraelectric $(\text{PbTiO}_3)_n/(\text{SrTiO}_3)_n$ superlattices [1]. This novel phase is stabilized by the interplay of lattice, charge, and orbital degrees of freedom. The vortices have a multi-order parameter state, characterized by both a toroidal moment and an axial component of the polarization [2]. This three-dimensional polarization topology suggests the possibility of a chiral state, reminiscent of complex spin textures such as magnetic skyrmions.

Resonant soft X-ray diffraction (RSXD) experiments in conjunction with second-principles simulations have shown that this vortex phase is indeed chiral [3]. RSXD studies were performed at the titanium $L_{3,2}$ edge, with the diffraction geometry aligned to observe satellite diffraction from the vortex ordering. By taking the difference of the diffraction intensity observed for left- and right-circularly polarized X-rays, the X-ray circular dichroism (XCD) is measured. The asymmetry of the XCD signal observed in this context is a hallmark of chirality in the superlattices. RSXD is so far the only technique capable of identifying the handedness of this material system, due to its sensitivity to chiral polar distortions of the TiO_6 octahedra with nm-scale periodicity. Furthermore, we have identified a promising mechanism for switching the handedness of these chiral superlattices. The multi-order parameter state could allow for coupling between the toroidal moment of a vortex and its ferroelectric-like axial component of polarization. Using this coupling, an applied electric field could be used to selectively control the chirality, switching vortices from their initial state to the chiral enantiomer.

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Strain Engineering of Phonon Modes in (111)-Oriented Perovskites

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Strain engineering is a valuable tool to control functional properties in perovskite oxides. It has e.g. been used to induce ferroelectricity in paraelectric SrTiO₃¹ and to control the rotations of the oxygen octahedra, considered essential for electronic and magnetic properties.² The control of functional properties can be related to a change of preferable phonon modes of the material due to the imposed strain state. Recent advances in thin film growth now enables atomic control synthesis of various crystalline facets including the trigonal oriented (111) facet, opening new vistas for strain control.

Here we present a density functional theory calculation study of response to strain applied in the (111)-plane. For (001)-strain, the phonon properties follow the established intuitive trends, increased out of plane rotation for compressive strain and in-plane rotations for tensile strain, fig. 1a. The response to strain in the (111)-plane is more complex. In fig. 1b the response for (111)-strained LaAlO₃ is shown, and the strain induced rotations of the oxygen octahedra is opposite as compared to (001)-strain. In addition, the in-plane octahedral rotations under (111)-strain can result in Goldstone-like modes.³

To better probe this we compare how (111)-strain affects phonon frequencies of rotational and polar modes for 20 different perovskites, including III-III, IV-II and V-I types. We show that the preferred phonon modes can be rationalized in terms of both the Goldschmidt tolerance factor and the formal oxidation states of the cations. Trends for coupling between (111)-strain and octahedral rotational modes, as well as polar modes, providing guidelines for rational design of (111)-oriented perovskite thin film systems will be presented.

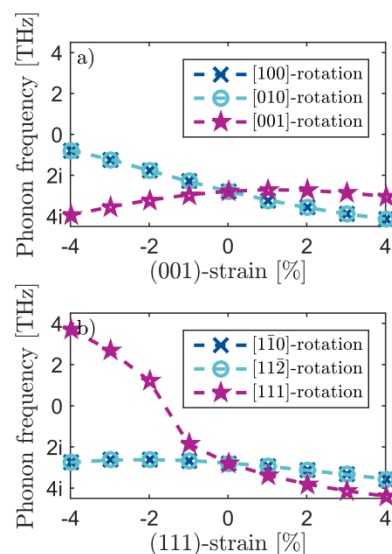


Fig 1. Rotational Phonon modes of a) (001)-oriented and b) (111)-oriented thin films.

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Reversible Modulation of Magnetic Anisotropy in $\text{Pb}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3/\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ Multiferroic Heterostructures

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Voltage control of magnetic anisotropy is critical for magnetoelectric (ME) random access memory device applications. Recently, $\text{Pb}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ (PZT)/ $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ (LSMO) heterostructures have gathered much attention due to large charge induced ME coupling in these heterostructures[1]. In this work, we report a reversible modulation of in-plane magnetic anisotropy energy (MAE) in 4 nm LSMO thin films via ferroelectric field effect induced charge doping facilitated by PZT top gate.

We have deposited high-quality PZT/LSMO heterostructures on (001) SrTiO_3 substrates via off-axis RF-magnetron sputtering. The ferroelectric field effect modulation in carrier density induced a 25 K change in metal-insulator transition temperatures (T_{MI}) in LSMO between the accumulation and depletion states. Below T_{MI} , the in-plane transverse resistivity of LSMO exhibits sharp field-symmetric jumps known as Giant Planar Hall effect(GPHE)[2]. In addition to the modulation of resistance jumps in GPHE due to polarization switching, we have also observed, increased pinning fields in the accumulation state, compared to the depletion state indicating a change in the magnetic energy landscape. We employed, planar Hall effect measurements (PHE) to study the in-plane magnetic anisotropy in PZT/LSMO heterostructures [2]. The magnetic anisotropy in LSMO is found to be biaxial with easy axes along $\langle 110 \rangle$ directions for both polarization states. The extracted biaxial anisotropy fields (H_i) from PHE measurements, showed an enhanced H_i in the accumulation state. Assuming a doping level change of 0.1 electron/Mn- due to the polarization switching [3], the estimated anisotropy energy densities are 1.1×10^5 erg/cm³ and 1.48×10^5 erg/cm³ in the depletion and accumulation states respectively, corresponding to 30% enhancement of MAE in the accumulation state, when compared to the depletion state. First principle density functional calculations performed for various Sr doping levels also show an increase in the MAE with an increase in the hole doping, agreeing well with our experimental observations. We attribute this enhancement in MAE to the modulation of orbital occupancy at the PZT/LSMO interface [4]. Our work provides new insights to study and control MAE in CMR oxide thin films for spintronic applications.

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Orbital Polarization in Negative Charge Transfer Ferrate Perovskites

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Controlling the electron orbital occupancy of transition metal oxides (TMOs) can lead to novel electronic and magnetic properties that may enable new electronic devices. Such orbital polarization (OP) can be induced via epitaxial strain due to the coupling between electronic and structural degrees of freedom inherent in TMOs. In systems with octahedral coordination, such as perovskites, biaxial strain induces OP in nominally e_g' systems by lowering the symmetry around the TM atom and thus removing the e_g orbital degeneracy. However, some transition metal oxides, such as nickelates and ferrates, have strongly hybridized TM $3d$ and O $2p$ orbitals, which is typically characterized by a negative charge transfer energy—the energy cost of transferring an electron from an O $2p$ orbital to a TM $3d$ orbital. This increased covalency can significantly alter e_g OP. The interplay between strong hybridization, orbital polarization, and strain is not well understood but such an understanding is needed to rationally design systems with large orbital polarization.

Here we measure X-ray linear dichroism (XLD) of epitaxially strained SrFeO_3 (SFO) and CaFeO_3 (CFO) films, which are nominally e_g' (d^4) systems, but are better described as $\alpha|d^4\rangle + \beta|d^5L\rangle$ owing to their negative charge transfer energy [1]. Strained SFO and CFO films were grown by oxygen plasma-assisted molecular beam epitaxy on various (001) single crystal substrates. X-ray diffraction and electrical transport measurements confirm that the films are epitaxially strained and fully oxidized [2]. XLD of the Fe L -edge shows preferential occupation of $d_{x^2-y^2}$ ($d_{3z^2-r^2}$) orbitals for tensile (compressive) strain, as would be expected from the strain-modified crystal field. By measuring X-ray absorption of the O K -edge and its pre-peak, which quantifies the ligand hole concentration, we show how strain affects the TM-O bond covalency. These results are compared to simulated spectra generated from a full-correlation, double cluster model that captures negative charge transfer, bond and charge disproportionation, and the necessary orbital degeneracies and Coulomb interactions. Taken together, these results provide further insight into strain-induced orbital polarization in strongly hybridized correlated oxides.

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Electrochemical and Mechanical Control of Metal-Insulator Transition in Epitaxial Vanadium Dioxides

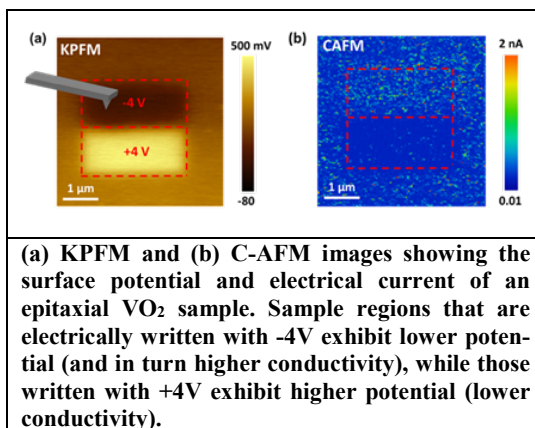
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Strongly correlated vanadium oxide (VO₂) is one of the most promising material exhibiting temperature-driven metal-insulator transition (MIT) in the vicinity of room temperature. Understanding electrochemical and mechanical properties of VO₂ is important to gain precise control over the conductivity of VO₂ thin film nanostructures by means of external stimuli, such as strain, temperature, and electric field. Previous studies have focused on exploring the influence of macroscopic mechanical strain and electrostatic gating on the MIT of VO₂. However, exploring such external influence at localized, microscopic length scales have been missing. In this work, we study electrochemical and mechanical modification of the MIT in VO₂ thin films epitaxially grown on (001) TiO₂ substrates, using Kelvin probe force microscopy (KPFM) and conducting-atomic force microscopy (C-AFM). We identified that the applied electric voltage and mechanical pressure through the SPM-tip could locally modulate VO₂ film's conductivity. Surface chemical potential and conductivity variations were measured as a function of temperature in the vicinity of MIT. Our results could be attributed to the local change in the oxygen stoichiometry in VO₂ films, and provide the basis for local electrochemical control of MIT in VO₂ thin films.



This work was supported by the Center for Predictive Simulation of Functional Materials, U.S. Department of Energy (DOE), Basic Energy Sciences (BES) Computational Materials Sciences Program (KPFM, AFM) and by the Materials Sciences and Engineering Division (synthesis). The use of atomic force microscopy was performed as a user project at the Center for Nanophase Materials Sciences, which is sponsored at ORNL by the Scientific User Facilities Division, BES, U.S. DOE.

Tunable Band Gaps in Digital Oxides with Layered Crystal Habits

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Many atomically layered oxide superlattices are constructed from three-dimensionally connected bulk perovskites. Here we examine the electronic structure of Ruddlesden-Popper (RP) oxides, focusing on (LaSr)AlO₄ and changes induced by A cation order. Recently the aluminate RP superlattice was predicted to show massive band gap variations, up to 200%, depending on the A cation ordering [1]. This variation was attributed to changes to the internal electrostatic potential and electric fields within the superlattice mediated by ionic relaxations. In this work we extend that concept, showing that longer period arrangements of the monoxide planes lead to greater control over the band gap and even its full collapse, i.e., a metal-insulator transition is observed. Density functional theory calculations are used to investigate the underlying mechanism of this band gap variation. We verified that the band gap control originates from the internal electric fields induced by the digital chemical order, which induces picoscale band bending; the electric field magnitude is mainly governed by the inequivalent charged monoxide layers afforded by the layered crystal habit. Its impact on the band gap variation is quantitatively compared with secondary contributions including charge transfer and ionic relaxations. We conclude by summarizing our findings into design principle principles for band gap control in layered oxides, whereby our formulated descriptor captures the layer-charge variation and also predicts changes in the electronic gap due to antisite defects and in other chemistries.

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Dimensionality-Driven Semimetal-Insulator Transition in Ultrathin Films of Spin-Orbit Coupled SrIrO_3

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The Ruddlesden-Popper phases $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ are discussed to exhibit a semimetal to insulator transition for the series $n=\infty, 2, 1$ layers of SrIrO_3 , where the $n=1$ member is a so-called spin-orbit driven Mott insulator [1-4]. Here the spin-orbit coupling is large enough to split overlapping, mixed bands into a manifold of narrow bands with well-defined effective total angular momentum J_{eff} . With lowering the dimensionality of the IrO_2 plane structure these bands become successively more susceptible to Mott localization since the coupling strength gets enhanced even though the local onsite Coulomb repulsion U is relatively small in $5d$ as compared to $3d$ transition metal oxides.

A systematic investigation of the $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$ series, however, is difficult since the bulk compounds are unstable in ambient pressure for $n \geq 3$ and thus extremely challenging to grow as single crystals. An alternative way to investigate the effect of dimensionality on the electronic properties is the fabrication of ultrathin SrIrO_3 films with varying thickness.

We have grown high-quality SrIrO_3 thin films on SrTiO_3 by pulsed laser deposition and investigated their electronic structure by soft x-ray angle resolved photoemission. Indeed, a semimetal-insulator transition is found as function of film thickness as evidenced by the opening of a gap at the chemical potential. We discuss the nature of the observed transition based on density-functional calculations including U (DFT+ U), taking the thickness-dependent octahedra rotation and tilting as observed by low-energy electron diffraction into account. While the inclusion of electron correlations and structural distortions is required in order to reproduce the electronic dispersions, DFT+ U can reproduce the insulating ground state only if (antiferro)magnetic order is considered. The phase transition appears as a complex, concerted action in which structural distortions, electron correlations, spin-dependent hopping enabled by the entanglement of orbital and spin wave functions as well as magnetic coupling all cooperate in driving the transition upon tuning the dimensionality.

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Strain control of cationic distribution in multiferroic $\text{Bi}_5\text{Ti}_3\text{FeO}_{15}$ films

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Strain engineering has been a powerful way to improve or to develop novel functionalities of materials by modulating the bond angle/length between atoms as well as inducing defects. Beyond these well-known mechanisms, a recent theoretic calculation predicted that strain engineering can even control the cation distribution of ions in the well-known Aurivillius multiferroic system, $\text{Bi}_5\text{Ti}_3\text{FeO}_{15}$ ($\text{Bi}_4\text{Ti}_3\text{O}_{12} + \text{BiFeO}_3$) [1]; while Ti and Fe ions are randomly distributed in bulk, Fe ions can preferentially locate at the inner or outer octahedral layers in the Aurivillius structure depending on the type of strain. To attest the prediction, we have fabricated $\text{Bi}_5\text{Ti}_3\text{FeO}_{15}$ epitaxial films on SrTiO_3 , LSAT, and LaAlO_3 substrate by using pulsed laser epitaxy. These substrates allow us to control both the sign and degree of strain ($-0.94 - 1.20\%$). Scanning transmission electron microscopy with energy dispersive x-ray spectroscopy revealed that Fe ions have site-preference when compressively strained, whereas no preferential positioning was observed in the tensile strained films. This experimental result supports well the theoretical prediction particularly for the case of compression, while further theoretical and experimental attempts are required for the tensile case. Effects of the site preference on magnetic and ferroelectric properties were investigated by using SQUID and second harmonic generation and will be discussed.

This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

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MBE grown Lutetium deficient $(\text{Lu}_{1-x}\text{FeO}_3)_m/(\text{Lu}_{1-y}\text{Mn}_{(1/3)}\text{Fe}_{(2/3)}\text{O}_3)_n$ superlattices

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Hexagonal rare earth ferrites and manganites have a variety of useful properties ranging from ferroelectricity, to ferromagnetism and antiferromagnetism. In addition, with their similar structures and lattice constants, they can be combined epitaxially in interesting ways. Molecular beam epitaxy gives a unique way to create materials that are not stable in bulk, or combine multiple structures to form superlattices that can have different properties from their layers individually. These techniques have been used to create Lutetium deficient $(\text{Lu}_{1-x}\text{FeO}_3)_m/(\text{Lu}_{1-y}\text{Mn}_{(1/3)}\text{Fe}_{(2/3)}\text{O}_3)_n$ superlattices by depositing Manganese and Iron without any additional Lutetium on top of stoichiometric LuFeO_3 . The Lutetium is pulled out of the LuFeO_3 layer into the $\text{Lu}(\text{MnFe})\text{O}_3$ layer. This creates a Lutetium deficiency not only in the $\text{Lu}(\text{MnFe})\text{O}_3$ layers, but also in the LuFeO_3 layers. Normally LuFeO_3 compensates for Lutetium deficiency by forming LuFe_2O_4 , but with this method no LuFe_2O_4 is created. The properties of these films are currently being investigated.

High-Throughput Screening of Transparent Conducting Oxides

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An investigation of the bandgap energies (for transparency) and effective masses (for mobility) of several current and new polymorphs of group-III sesquioxides (M_2O_3 , where $M = Al, Ga, \text{ and } In$) for screening as wide bandgap semiconductors will be presented. Through an examination of these two properties for many different lattice structures, the bandgap energies for Al_2O_3 , Ga_2O_3 and In_2O_3 vary by *ca.* 2 eV, 1 eV, and 0.5 eV, respectively. For the effective masses, an opposite trend is computed, with the largest change found for In_2O_3 , then Ga_2O_3 , and to a lesser degree the Al_2O_3 polymorphs. The underlying physical origins of this variation are detailed and represent an important step towards understanding the measured transparency and efficient electron mobility in In_2O_3 and Ga_2O_3 . With an understanding of the pristine phases of group-III sesquioxides, we then systematically examine the thermodynamics of ordering and phase separation of ternary and quaternary $(In_xGa_yAl_z)_2O_3$ (where $x+y+z=2$) materials using DFT-based cluster expansion models combined with fast stochastic optimization techniques (nested sampling) to efficiently search (meta)stable configurations for many lattice types. This approach allows for a quantitative theoretical phase diagram over the full composition range for several crystalline polymorphs experimentally identified for Al_2O_3 , Ga_2O_3 and In_2O_3 . With an extensive search over configurational space, statistical learning is performed for the bandgaps and stabilities to identify structure-property relationships between the targeted properties (e.g., optical transparency) and the fundamental chemical and physical parameters that control these properties.

Observation of Topological Hall Effect in Highly-Conductive SrRuO₃ Thin Films

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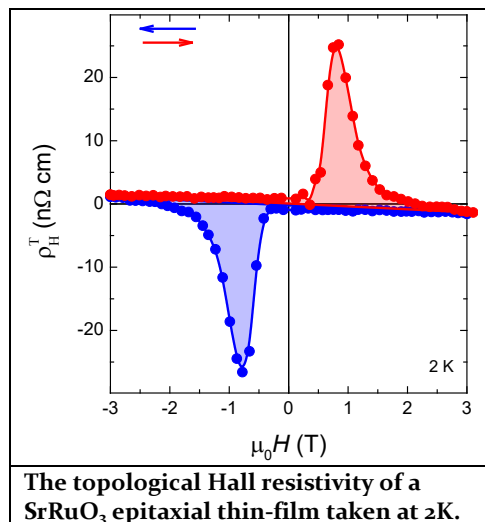
Skyrmions are topologically protected particles characterized by a chiral spin structure and associated with a topological integer which cannot be altered by a continuous deformation of the field configuration. Moreover, since magnetic skyrmions comprise many spins, they are expected to be robust under thermal and quantum fluctuations making them ideal platforms for investigating fundamental sciences and proof-of-concept devices.

The chirality which is necessary for magnetic skyrmion formation is a result of the Dzyaloshinskii-Moriya interaction (DMI) which requires both a strong spin-orbit interaction (SOI) as well as broken inversion symmetry. Therefore, one would expect to find magnetic skyrmions abundant in 4d and 5d transition metal oxide thin films which exhibit strong SOI and inherently lack inversion symmetry. Unfortunately however, there have been little reports of magnetic skyrmions in complex-oxide systems. Only in the last two years have reports begun to emerge in systems such as EuO_{1-x}[Ref. 1] and SrRuO₃-SrIrO₃[Ref. 2], but a clear understanding of the nature of magnetic skyrmion formation is not yet realized.

We have observed the topological Hall effect (THE), presumably due to magnetic skyrmions, in highly-conductive SrRuO₃ epitaxial thin-films grown on GdScO₃ substrates. Note that this system does not have the added complexity of oxygen stoichiometry or hetero-interfaces of materials with large SOI. We have separated the topological contribution to the Hall resistivity from the ordinary and anomalous terms by rotating the sample with respect to the field. We have observed that THE vanishes leaving the ordinary and anomalous terms intact at some critical angles. Our results show that it is possible to induce the formation of magnetic skyrmions in complex-oxide thin films without the need for complicated stoichiometry and/or multilayers. By eliminating these complexities we are able to more closely examine how the topological properties of materials and skyrmions are affected by the DMI. This work will ultimately pave the way for functional oxide electronics that take advantage of the unique properties of magnetic skyrmions.

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On the Universality of Electron Mobility in $\text{LaAlO}_3/\text{SrTiO}_3$ and bulk SrTiO_3

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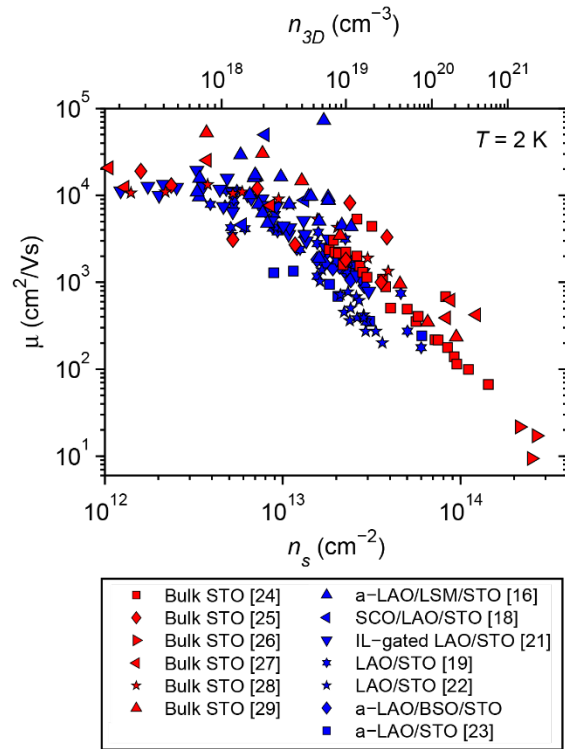
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Metallic $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) interfaces attract enormous attention, but the relationship between the electron mobility and the sheet electron density, n_s , is poorly understood. Here we derive a simple expression for the three-dimensional electron density near the interface, n_{3D} , as a function of n_s and find that the mobility for LAO/STO-based interfaces depends on n_{3D} in the same way as it does for bulk doped STO. It is known that undoped bulk STO is strongly compensated with $N \approx 5 \times 10^{18} \text{ cm}^{-3}$ background donors and acceptors. In intentionally doped bulk STO with a concentration of electrons $n_{3D} < N$ background impurities determine the electron scattering. Thus, when $n_{3D} < N$ it is natural to see in LAO/STO the same mobility as in the bulk. On the other hand, in the bulk samples with $n_{3D} > N$ the mobility is lowered because scattering happens on n_{3D} intentionally introduced donors. For LAO/STO the polar catastrophe, which provides electrons, is not supposed to provide an equal number of random donors and the mobility should consequently be larger than what is observed. The fact that the mobility is still the same as bulk implies that for LAO/STO the polar catastrophe model should be modified.



The dependence of the low-temperature electron mobility, μ , on the sheet carrier density, n_s , as well as corresponding bulk carrier density, n_{3D} , for bulk STO (red markers) and LAO/STO-based interfaces (blue markers) studied by different groups.

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Tuning ferromagnetism at the $\text{LaNiO}_3/\text{CaMnO}_3$ interface via crystallographic symmetry mismatch

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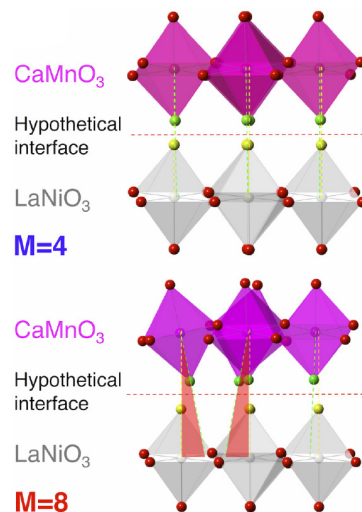
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Creating and tuning ferromagnetism at the interfaces between two non-ferromagnetic materials is of fundamental and technological interest. Remarkably, in the CaMnO_3 (CMO) perovskite that is normally antiferromagnetic, a fractional charge transfer of the order of 0.07 e-/Mn induces a canted antiferromagnetic state in the interfacial Mn.[1] In this way, at the interface, a magnetic ground state is stabilized by a delicate balance between ferromagnetic double exchange and antiferromagnetic super exchange interactions which is highly sensitive to the local structural distortions. Therefore, creating subtle structural modifications at the CMO interface can lead to large changes in magnetization. For example, one can modify interfacial ferromagnetism by a factor of two by using a second similar perovskite oxide but with different crystallographic orientation thus inducing a symmetry mismatch at the interface via dissimilar octahedral rotations.[2]

In this work we combined two perovskites with distinct crystallographic symmetries: the orthorhombic CaMnO_3 perovskite exhibiting the $a^-a^-c^+$ octahedral rotational pattern and the rhombohedral LaNiO_3 (LNO) exhibiting $a^-a^-a^-$ rotations. We show that in LNO/CMO superlattices we can control the octahedral connectivity across the LNO/CMO interface by changing the LNO and CMO layer thicknesses as well as their relative ratio and this way effectively alter the balance between ferromagnetic and antiferromagnetic interactions in the interfacial CMO layer. We have found that in LNO/CMO superlattices, the interfacial ferromagnetism can be modulated by a factor of three. The origin of the interfacial ferromagnetic variation is directly related to changes in Mn-O-Ni and Mn-O-Mn bond angles and bond lengths. The results demonstrate a new path for creating and controlling magnetic state via octahedral connectivity at the interface between two non-ferromagnetic materials with distinct crystallographic symmetries.



M=8
Hypothetical interfacial alignment for $M=4$ and $M=8$ superlattices demonstrating $M=8$ mismatch due to in-plane orthorhombic orientation.

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Giant topological Hall effect from magnetic skyrmion bubbles in correlated manganite thin films

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CaMnO₃ is a charge-transfer insulator pertaining to the widely studied family of colossal magnetoresistance manganites. The peculiarity of this oxide is that the chemical substitution of Ca²⁺ by Ce⁴⁺ gives two electrons to the system and raises the Fermi level, inducing a transition to a metallic and weakly ferromagnetic phase at 5% Ce concentration in bulk [1] and only 2% in compressively strained thin films grown on YAlO₃(001) substrates [2]. The low critical electron doping required to reach a metallic state makes this compound very interesting as a channel material for field effect studies [3] [4], and to explore the interplay between electron doping, correlation effects and disorder in perovskite oxides.

This lightly-doped material features a competition between super-exchange, double-exchange and Dzyaloshinskii-Moriya interaction which stabilize a highly non-coplanar G_xA_yF_z spin structure. In addition, on YAlO₃(001) substrates the magnetoelastic anisotropy imposes a perpendicular easy magnetization axis. This combination of complex spin configurations and a perpendicular easy axis makes this compound attractive to probe for sought-after topological effects, such as magnetic skyrmions, in correlated oxides [5].

In this presentation we will report [6] the presence of an anomalous Hall effect (AHE) and, secondly, of a topological-like Hall signal (THE) in these manganite thin films. Remarkably, both signals are very large and present up to low temperatures, and we show that this behavior for the AHE is due to the highly non-coplanar spin structure of the compound. Magnetic force microscopy reveals that the magnetization reversal nucleates magnetic bubbles, whose density directly relates to the THE signal. The magnitude of the THE is much higher than both any previously reported and what would expect from such a skyrmion bubble density. We point out how this, in addition to the relationship between the THE and AHE in the metallic range of Ce doping, can be ascribed to the strong correlations close to the metal-to-insulator transition.

This work received financial support from ERC MINT, ANR FERROMON and Labex Nanosacalay.

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Activating and stabilizing oxides in electrochemical devices: creating new Oxide-Electrolyte/Oxygen Interfaces in SrIrO₃/DyScO₃ films

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The design of efficient and stable metal-oxide catalysts for oxygen evolution reaction (OER) is crucial for the development of electrochemical energy conversion devices, such as electrolyzers and metal–air batteries^{1,2}. Currently, such design is challenged and limited by the instability of metal oxides during the OER, since OER processes involve elemental exchange between the oxide surface and the electrolyte, as well as the gas environment. This often leads to dissolution of oxides, resulting in an undesired decrease of performance^{3,4}. Here, we examined the model system of epitaxial SrIrO₃ films grown on DyScO₃ (110) substrates, combining electrochemical measurements, synchrotron X-ray scattering, and X-ray spectroscopic studies to understand the activation and stabilization of OER catalysts in acidic electrolytes. It was found that besides electrochemical activation, there is an evolution into stable oxide-electrolyte/oxygen interfaces at the top layers with a thickness of 6 unit cells. Theoretical calculations show that as compared with pristine SrIrO₃ oxide, these interfaces demonstrate greatly facilitated exchange of lattice oxygen in the reconstructed frameworks, oxygen molecules, and water, protecting the bulk oxide underneath via the formation of a diffusion barrier at interfaces. Our results illustrate the critical role of elemental etching on surface structures and lattice oxygen activation for enhanced OER, offering a new paradigm for balancing the activity and stability of oxides for various electrochemical device applications.

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Origin of interface ferromagnetism in $\text{LaMnO}_3/\text{SrTiO}_3$ heterostructures

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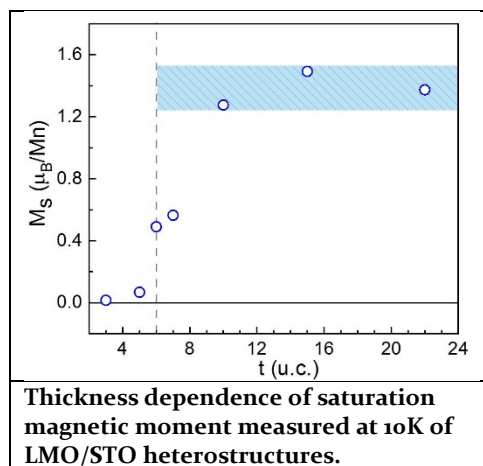
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Thickness control within unit cell (uc)-level has been achieved by the development of modern techniques for oxide thin films. Atomically sharp oxide heterostructures exhibit numerous novel properties which are absent in the bulk states¹⁻³. A prominent example is the remarkably sharp transition from insulating state to metallic behavior of the $\text{LaAlO}_3/\text{SrTiO}_3$ heterostructure when the thickness of LaAlO_3 is higher than 3 uc. Meanwhile, here we demonstrate an emergent transition at the $\text{LaMnO}_3/\text{SrTiO}_3$ (LMO/STO) interfaces: When the thickness of LMO is 6 uc or more, the interface becomes ferromagnetic. However, the underlying physics behind the LMO/STO interfaces showing thickness-dependent sharp magnetic transition still remains elusive. Herein, by using X-ray magnetic circular dichroism and electron energy-loss spectroscopy analysis with atomic resolution in an aberration-corrected scanning transmission electron microscopy, we discuss the origin of ferromagnetism at the LMO/STO interface, which is attributed to the common existence of “dead-layer” at the heterostructures⁴.



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Stoichiometry, Strain Relaxation and Electronic Disorder in Hybrid MBE Grown La-doped SrSnO₃ Films

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Materials possessing wide bandgap and high room temperature mobility are needed for high power transistor application and for transparent electrode function. Recently, doped BaSnO₃ (BSO) has generated a lot of excitement owing to a bandgap of ~ 3 eV and high room temperature (RT) mobility. Doped SrSnO₃ (SSO), on the other hand, has wider bandgap, 3.9-4.5 eV and also possesses lattice matching (orthorhombic, *Pnma*, 4.04 Å) with many commercially available substrates. Compared to BSO, SSO is however much less explored, perhaps due to the difficulties associated with the doping. Recent attempt to grow conducting SSO films by molecular beam epitaxy (MBE) has been largely unsuccessful. Growth explorations of doped SSO films using pulsed laser deposition have yielded a RT mobility value 40 cm²V⁻¹s⁻¹ at a carrier density of 9×10^{19} cm⁻³.

In this study, we will present a detailed study of MBE growth, strain relaxation and electronic transport in La-doped SSO films grown on a range of substrates LaAlO₃(001), SrTiO₃(001) and PrScO₃(110), and GdScO₃(110) by hybrid MBE. Reflection high-energy electron diffraction showed a layer-by-layer growth mode. High-resolution XRD revealed the out of plane lattice parameter of 4.04 Å for a 54 nm-thick SSO films grown on LaAlO₃(001) at a substrate temperature of 900 °C in the presence of oxygen rf plasma (operated at 250W and at oxygen pressure of 5×10^{-6} Torr), indicating these films were completely relaxed. Films on PrScO₃ (-0.2%, compressive) and GdScO₃ (-1.6%) remained completely strained up to 50 nm. Cation stoichiometry was optimized using a combination of high-resolution XRD and Rutherford backscattering spectrometry.

Electronic transport revealed a RT mobility of 55 cm²V⁻¹s⁻¹ at a carrier density of 5.6×10^{19} cm⁻³ for La-doped SSO films on GdScO₃(110) whereas much smaller mobility ~ 20 cm²V⁻¹s⁻¹ was obtained in relaxed films on LaAlO₃(001) at same doping levels revealing an important role of dislocation on electronic transport. Temperature dependent transport analysis further revealed a crossover from 2D to 3D weak localization at low temperatures with increasing amount of dislocations. Strong localization of carrier is observed for increasing amount of non-stoichiometry with the transport behavior governed by the Mott 3D variable range hopping (VRH) for very non-stoichiometric films. In nutshell, using a combination of detailed MBE growth, XRD, STEM, spectroscopy and temperature-dependent electronic transport study, we will discuss the important role of specific defects such as dislocation and non-stoichiometry on electronic transport in doped SSO films.

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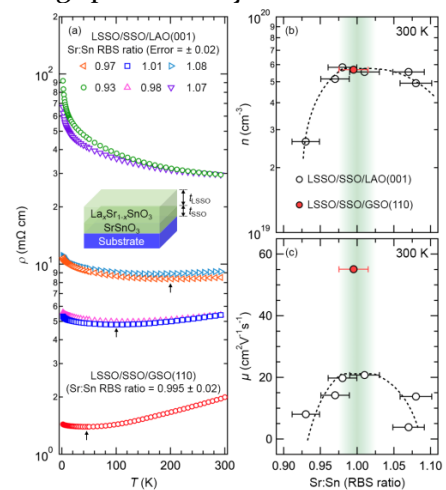


Figure 1: electronic transport of La-doped SSO films on LaAlO₃(001) and GaScO₃(110)

Digital modulation of nickel valence in cuprate-nickelate heterostructures

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The past years have seen sophisticated approaches to design novel materials aiming to engineer their properties.^[1] Charge doping is a key tuning parameter. But even with δ -doping, the dopant cannot be confined to one atomic layer due to the diffusion of dopant in growth direction.^[2] Thus, effects of charge and disorder cannot be separated. Here, we suggest doping through insertion of a positively charged layer, LaO^+ , which allows for a digital modulation of the nickel valence in cuprate-nickelate heterostructures.

We use atomic layer-by-layer, ozone assisted molecular beam epitaxy to grow structures with the chemical formula $[(\text{La}_2\text{CuO}_4)_n\text{LaO}(\text{LaNiO}_3)_m]_l$ and a structure as shown in Fig. 1. X-ray absorption measurements confirm that the charge provided by the LaO^+ layer is exclusively accommodated in the nickelate layers, leaving the cuprate layers as inert separating layers. The electrons reside mostly in the out-of-plane, z^2 -orbitals and the valence state of nickel is modulated within the nickelate stack. DFT simulations show that a gap opens in the interfacial nickelate layers which is supported by electrical transport measurements.

The insertion of LaO^+ layers shows a way of charge doping without introducing disorder. Moreover, if one LaO^+ layer at the bottom of a nickelate layer is replaced by SrO , the diffusion in growth direction will be blocked by the nickelate layers which can only be hole-doped in very high pressures not available for MBE growth and true δ -doping could be feasible.

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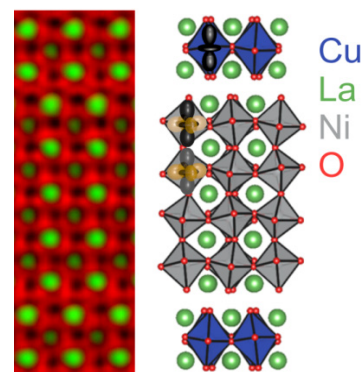


Figure 1. Combined high-angle annular dark field and annular bright field STEM image (left) and structure obtained from DFT simulations (right) of $[(\text{La}_2\text{CuO}_4)_3\text{LaO}(\text{LaNiO}_3)_4]_6$. La atoms appear bright green, Ni and Cu atoms dark green and oxygen atoms appear black in the STEM image. The orbitals added to the DFT structure represent the distribution of the electrons residing mainly in the z^2 -orbitals.

Magnetic Anisotropy of NiCo₂O₄ thin films

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NiCo₂O₄ is a conducting oxide with a spinel structure. It is promising for the application of energy storage devices as a high-efficiency electrode material.¹ The origin of the conductivity is believed to be related to the mixed valent Co and Ni ions. A ferrimagnetic order above room temperature has been observed in NiCo₂O₄ with the Co moments anti-aligned to the Ni moments². Due to the contribution of magnetization from magnetic ions of different species in different local environment and with different valences, the magnetic anisotropy in NiCo₂O₄ is expected to be complex and has not been fully addressed.

In this work, we have studied the magnetic anisotropy of the NiCo₂O₄ epitaxial thin films grown on MgAl₂O₄ substrates using the magneto-optical Kerr effect (MOKE) for various crystal orientations and thickness. For the (111)-oriented films, the magnetic easy axis show a three-fold rotational symmetry which is consistent with the crystalline symmetry. The easy-axis rotates toward the out-of-plane direction when the film thickness is reduced. For the (001)-oriented films, an easy axis lying mostly out-of-plane was observed with a uniaxial symmetry; the direction of the easy axis is not aligned with any high symmetry direction of the film. The implication of these observations on the single-ion magnetic anisotropy will be discussed.

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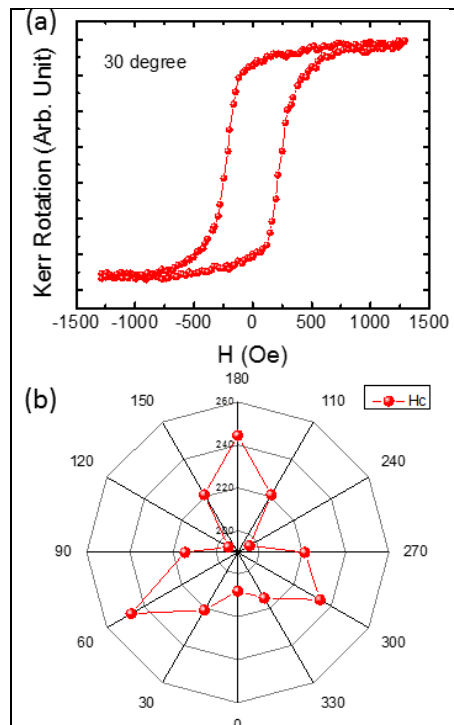


Figure 1. (a) Hysteresis loop of NiCo₂O₄ (111) films measured using the magneto-optical Kerr effect. (b) Angular dependence of the Coercivity of the NiCo₂O₄ films.

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Enhanced water oxidation activities of α -Fe₂O₃ photoanode by introducing Co and P dopants

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Hematite (α -Fe₂O₃) is a promising candidate for the photoelectrochemical (PEC) water oxidation photoanode. However, owing to polaron hopping conduction through a Fe²⁺/Fe³⁺ valence alternation on spatially localized 3d orbitals, the intrinsic low carrier mobility and short diffusion length of α -Fe₂O₃ result in poor conductivity, which greatly limits its application. Doping is an effective approach to improve the performance. In this work, we successfully doped Co and P into α -Fe₂O₃ by a simple chemical method. The photocurrent density (at 1.23 V vs. RHE) was enhanced by a factor of 12.5 and 20.6 times in the P- and Co- doped hematite samples, respectively. Roles of Co and P dopant in the enhanced PEC water oxidation activities of hematite were investigated by utilizing scanning electron microscopy, transmission electron microscopy, elemental analysis, X-ray diffraction. It has been revealed that Co dopant effectively decreases the flat band potential, and thus promotes the charge transfer at the photoanode/electrolyte interface. Moreover, Co dopant causes red-shift of the absorption edges, resulting in small optical band gap. P dopant is helpful to increase the donor density and improve the electric conductivity of hematite photoanode. The distinctive roles of Co and P dopants suggested us a feasible approach by Co and P co-doping to further enhance PEC performance of hematite, which will be studied in our future work.

Epitaxial SrTiO₃/BaSnO₃ Heterostructure MOSFETs

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La doped BaSnO₃ (La:BSO) has been considered to be of great potential for optoelectronics and transparent electronics application due to its high room temperature mobility, optical transmission and thermal stability¹⁻³. Here we report the fabrication of n-type depletion mode metal-oxide-semiconductor field effect transistors (MOSFETs) based on epitaxial SrTiO₃/BaSnO₃ (STO/BSO) heterostructures. Undoped BSO buffer layer, La:BSO active channel layer and STO gate dielectric layer were all deposited epitaxially on STO substrate using hybrid molecular beam epitaxy technique. MOSFETs with 50 μm gate-length achieved a maximum drain current density of 16mA/mm as shown in Fig. 1. This value is comparable to the highest reported values of BSO-based FETs⁴⁻⁸, indicating promising applications in power electronics. The maximum room temperature field-effect mobility of this device is around 60cm²V⁻¹s⁻¹. Compared with BSO/HfO₂ based devices, MOSFETs based on BSO/STO heterostructures showed superior performances, which likely originated from better interface quality owing to the epitaxial growth. We will also present detailed study of interface trap density, and detailed FET device characteristics using shorter gate length device architectures.

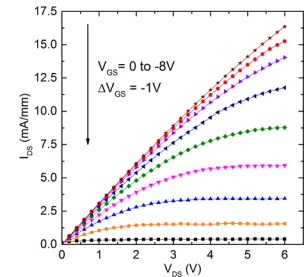


Fig. 1: Output characteristics of the 50 μm gate-length MOSFET based on BSO/STO heterostructure. The gate voltage was varied from $V_{GS}=0\text{V}$ to $V_{GS}=-8\text{V}$ with 1V interval.

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Two-dimensional electron liquid states at the K-adsorbed anatase-TiO₂ (001) surface

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The techniques to control the low-dimensional electronic states appearing at the surface and interface of oxide semiconductors have attracted huge attention for designing the future quantum electronic devices. Recently, the formation of two-dimensional electron liquid (2DEL) states has been reported at the surface of anatase titanium dioxide (a-TiO₂) [1]. The 2DEL is generated by irradiation of strong vacuum-ultraviolet light which induces oxygen vacancies near the surface. However, owing to the difficulty to control the distribution of the oxygen vacancies in depth, the dimensionality of the electronic states generated by photoirradiation remains a controversial issue [1,2]. Therefore, in this study, we have proposed an alternative route to address the 2DEL states: we adsorbed K atoms on the (001) surface of a-TiO₂ [3] and investigated the formed 2DEL states by *in situ* angle-resolved photoelectron spectroscopy (ARPES).

Figure 1(a) shows the Fermi surface (FS) determined by *in situ* ARPES at the bare and K-adsorbed a-TiO₂ (001) surfaces. The evolution of FS volume by K adsorption indicates that electrons are doped into a-TiO₂ and accumulated in the surface region as in the case of the field-effect-transistor structure. To investigate the dimensionality of the generated electronic states, an ARPES image is taken along the cut A as shown in Fig. 1(b). The clear sub-band structures are observed, indicating that the electronic structures formed at the surface have a two-dimensional character. The simulation based on the wedge-shaped potential approximation suggests that the doped-electrons are confined within a few nm from the surface.

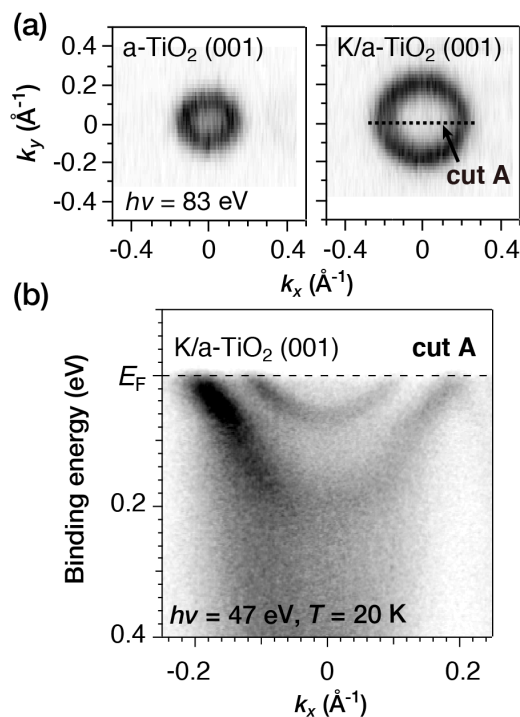


Fig. 1 (a) Evolution of the FS of a-TiO₂(001) surface by K adsorption. (b) ARPES intensity plot taken along the cut A in (a).

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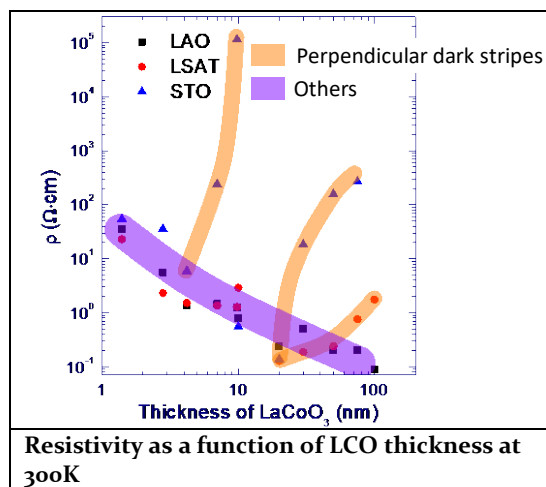
Conductivity and Microstructure of $\text{LaCoO}_{3-\delta}$ Thin Films

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Conductivity and microstructure of the varied thickness $\text{LaCoO}_{3-\delta}$ thin films have been systematically studied. Dark stripes of different orientations were observed in the $\text{LaCoO}_{3-\delta}$ films by scanning transmission electron microscopy, including the ones perpendicular to the interface in thick films on SrTiO_3 substrate, the ones parallel to the interface in thick films on LaAlO_3 substrate. But none is observed in thin $\text{LaCoO}_{3-\delta}$ films.^[1] The dark stripes are the superstructure formed by oxygen vacancies, which leads to the appearance of Co^{2+} due to charge compensation in $\text{LaCoO}_{3-\delta}$. The Co^{2+} is in high spin state and Co^{3+} in low spin state in $\text{LaCoO}_{3-\delta}$ films.^[2] Due to spin-state blockade^[3], the electron hopping from Co^{2+} (high spin state) to Co^{3+} (low spin state) is considerably suppressed. The orientations of the stripe have different influence on the conductivity of the film. The dark stripes perpendicular to interface will suppress the conductivity due to the inevitably passing through of the stripes for the transport of the electrons in the film. However on the contrary, the dark stripes parallel to interface has little effect on the conductivity. This work provides a new route to tune electrical conductivity by atomic-scale engineering in complex oxide heterojunctions.



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First-Principles study of the Metallicity of ZnO Surface

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Zinc Oxide (ZnO) surfaces are widely used in many applications such as catalysis, bio-sensing and solar cells¹⁻². These surfaces are, in many cases, terminated by hydroxyl groups. Experimental study by our collaborators has revealed³ a transition of ZnO electronic properties from semiconducting to metallic as the hydroxyl coverage increased to approximately 80%. The reason of this transition and main contribution to the metallicity of the surface is not well understood yet.

In our work, we performed first-principles calculations based on density functional theory for ZnO (10 $\bar{1}$ 0) surface with different amount of hydroxyl coverage, ranging from bare surface to fully covered surface. Full relaxation has been performed on these surfaces and we calculated the electronic band structure to verify the experimental finding of this semiconductor-metal transition. We then calculated the density of states of the surface to study the cause of this metallicity.

TABLE 1 SUMMARY OF SIMULATION RESULTS FOR DIFFERENT COVERAGES.

Coverage(monolayer)	0	0.25	0.5	0.75	0.83	0.875	1
Band gap (eV)	0.75	0.70	0.68	0.95	0.80	0.78	0
Surface O 2p state	No	Localized	Localized	No	Localized	Localized	Dispersive
Metallic surface (Simu.)	No	No	No	No	No	No	Yes
Metallic surface (Expt.)	No	No	No	No	Yes	Yes	Yes

Table I summarizes several key findings. First we found that the fully covered surface does not have a visible band gap, which means this surface is indeed metallic and there is a transition from semiconducting to metallic surface. Second, we found that the states near valence band maximum are oxygen 2p states. For metallic surface, these states are dispersive surface states and dominate the energy range across 1 eV below valence band maximum. Besides, by plotting site-projected density of states, we show that the main contribution to the surface states comes from oxygen atoms in the hydroxyl groups bonded to Zn atoms. Our simulation successfully verified the experimental finding of ZnO metallic surface and provided in-depth physical explanation to it. This work is support by National Science Foundation (DMR-1555153) and Blue Waters sustained-petascale computing project, supported by the National Science Foundation (awards OCI-0725070 and ACI-1238993). This project is also partially funded by the Computational Science and Engineering fellowship program in University of Illinois, Urbana-Champaign.

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Understanding Structural Basis of Ionic Electrolyte Gating on Oxide Heterostructures

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Due to unique fundamental behaviors of complex oxides, such as the subtle balance between competing electronic and magnetic phases, and their sensitivity to defects and doping, electric fields can be used to craft electronic order, modify chemistry, apply strain, and manipulate spin-orbit couplings (e.g. in the 5d oxides). This presents a promising opportunity to create novel functionalities, in principle, enabling device concepts that go far beyond what conventional semiconductor physics allows (i.e. adopting a scheme that emulates the neuron-circuit in the human brain). In particular, the very high charge density induced by an electric double layer (EDL) formed at an electrochemical solid-liquid interface has recently been used to induce or “gate” exotic phase transitions, therefore electronic ground states of strongly correlated oxides in the interfacial region, via the subtle interplay of electrostatic (electronic phenomena) and chemical redox effects (field-driven ionic motion). It is highly expected that leveraging ionic electrolyte gating would still be fertile ground for exploration in a broad range of oxides that exhibit novel functionalities.

In this talk, we will present our recent *in-situ* and real-time X-ray study to make fundamental understanding of structural basis (e.g. reconstruction and evolution) during EDL gating on various complex oxide heterostructures. The experimental findings from X-ray investigations illustrate distinct but contrasting structural responses to EDL field manipulation in these representative systems although most exhibit drastic modulation on transport due to the field effect. The EDL gating on underdoped cuprate heterostructures displayed a unique response mechanism, namely, that the strong electric fields can reversibly displace but not knock-off anions inside a unit cell while the overall lattice structure remains unchanged.¹ In contrast, the structural evolution in nickelates/ruthenate/tungstate heterostructures during EDL gating was generally consistent with controlling a metal-insulator transition by manipulating oxygen vacancies via redox chemistry.² More interesting to note that shuttling oxygen vacancies could lead to emergent controllability on structural motif of complex oxides, such as creating metastable brownmiller-phase and dynamic tuning oxygen octahedral rotation.

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Electron Doping of the Parent Cuprate La_2CuO_4 without Cation Substitution

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In quantum many-body systems, exotic phenomena are typically realized through carrier doping. One example is in the cuprates, where high- T_c superconductivity arises via doping of a Mott insulating parent state. To date, cation substitution has been the main method for doping carriers into these compounds, and is the only known method by which electron doping can be achieved in these materials. We report electron doping without cation substitution in epitaxially stabilized thin films of La_2CuO_4 grown via reactive oxide molecular-beam epitaxy. We use angle-resolved photoemission spectroscopy to measure the band structure and Fermi surface and directly determine the carrier concentration. We propose oxygen vacancies as the most likely source of doping in these materials. Our results suggest a new avenue towards electron doping with lower disorder in the cuprates and could lead to a more detailed experimental understanding of their properties.

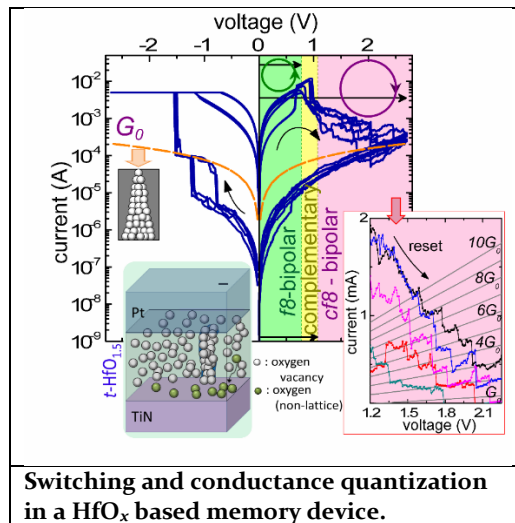
Control of Switching Modes and Conductance Quantization in Oxygen Engineered HfO_x based Memristive Devices

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Hafnium oxide (HfO_x) based memristive devices have tremendous potential as non-volatile resistive random access memory (RRAM) and in neuromorphic electronics. Despite its seemingly simple two-terminal structure, myriad of RRAM devices reported in the rapidly growing literature exhibit rather complex resistive switching behaviors. Using Pt/ HfO_x /TiN based metal-insulator-metal structures as model systems, we show that, a well-controlled oxygen stoichiometry governs the filament formation and the occurrence of multiple switching modes. The oxygen vacancy concentration is found to be the key factor in manipulating the balance between electric field and Joule heating during formation, rupture (reset), and reformation (set) of the conductive filaments in the dielectric. In addition, the engineering of oxygen vacancies stabilizes atomic size filament constrictions exhibiting integer and half-integer conductance quantization at room-temperature during set and reset. Identifying the materials conditions of different switching modes and conductance quantization contributes to a unified switching model correlating structural and functional properties of RRAM materials. The possibility to engineer the oxygen stoichiometry in HfO_x will allow creating quantum point contacts with multiple conductance quanta as a first step towards multi-level memristive quantum devices.



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Ferromagnetism and spin-dependent transport at a complex oxide interface

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Interfacial oxide systems have garnered a lot of attention after the discovery of a conducting interface between two insulating oxides LaAlO_3 and SrTiO_3 . This interfacial system has been shown to have two possible ground states, superconducting or ferromagnetic. Here we show via magneto-transport measurement at low temperatures [1] that the MBE-grown polar/non-polar $\text{NdTiO}_3/\text{SrTiO}_3$ (NTO/STO) interface hosts a ferromagnetic state.

We observe a very large negative magneto-resistance ratio (MR) of up to -95% at 150 mK . The amplitude of the negative MR decreases with increasing temperature and acquires a positive curvature above $\sim 4\text{ K}$ (Fig. 1a). By modeling electron transport at the interface using spin-dependent hopping, we obtain excellent quantitative agreement with the data over more than an order of magnitude in temperature (Fig. 1a). We find that the MR depends exponentially on the temperature (T) and magnetization (M) of the sample, following $MR \propto \text{Exp}[-\alpha(M/M_s)^2/k_B T]$, where α characterizes the spin-dependent energy cost of hopping. The system develops superparamagnetism below $\sim 4\text{ K}$, Fig. 1b. Above 1.5 K , all the superparamagnetic moments are above their blocking temperature and hence thermally fluctuate. Below 1.5 K the larger moments are blocked, leading to a hysteretic MR .

Furthermore, we show that at low temperatures time-dependent MR measurements are necessary to distinguish between magnetic effects intrinsic to the sample and extrinsic effects such as heating, which can also produce a hysteretic signal.

We conclude by discussing possible microscopic mechanisms that could lead to the formation of localized magnetic moments at the NTO/STO interface. Oxygen vacancies, dislocations or substrate impurities do not play an important role in determining the electronic properties of this system [2,3].

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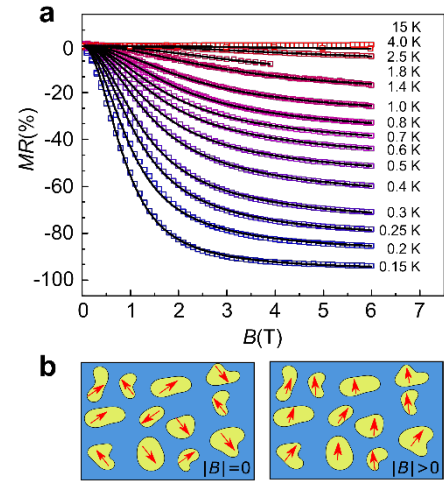


Fig. 1 (a) MR ratio of $\text{STO}(40\text{ uc})/\text{NTO}(4\text{ uc})/\text{LSAT}$ sample at different temperatures. The black lines are fits to a model based on spin dependent hopping. **(b)** Schematic showing arrangement of superparamagnetic islands at zero magnetic field (left panel) and finite field (right panel).

Metal-Insulator Transition in CaVO_3 Thin Films from DFT+DMFT

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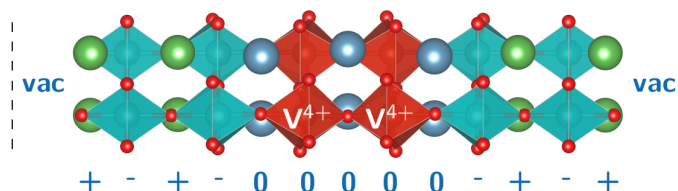
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The wide variety of interesting phenomena and emerging functionalities of complex oxide thin films and heterostructures is generally determined by a large number of different factors, such as e.g. substrate-induced epitaxial strain, dimensional confinement, interface-related effects, or defects.

Here, we investigate the effects of epitaxial strain, dimensional confinement, as well as interface and surface effects on the electronic properties of the correlated metal CaVO_3 using a combination of density functional theory (DFT) and dynamical mean-field theory (DMFT). We show that tensile epitaxial strain can induce a metal-insulator transition in CaVO_3 , and we demonstrate that this strain effect cooperates with a similar tendency originating from the finite thickness of the thin film, consistent with recent experimental observations [1].

However, the latter effect is quantitatively only relevant in the ultra-thin limit (a few unit cells) and does not facilitate a metal-insulator transition in thicker unstrained films. Furthermore, we also address the influence of a substrate-film interface on the structural and electronic properties of the film in both $\text{CaVO}_3/\text{LaAlO}_3$ (see Fig. 1) and $\text{CaVO}_3/\text{SrTiO}_3$ heterostructures, and we show that, while correlation effects are strongly enhanced at the CaVO_3 /vacuum surface, there is only a moderate enhancement at the heterostructure interfaces.

This work was supported by ETH Zurich and the Swiss National Science Foundation through NCCR-MARVEL.



Crystal structure, nominal charge per layer and nominal Vanadium valence for a thin $\text{CaVO}_3/\text{LaAlO}_3$ slab in vacuum. The dashed lines symbolize the cell boundaries along the c -direction.

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In-gap features in superconducting LaAlO₃ - SrTiO₃ - interfaces observed by tunneling spectroscopy

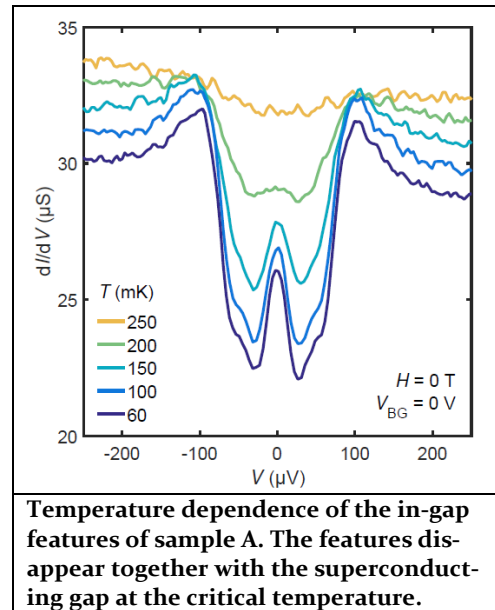
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We identified quasiparticle states at well-defined energies inside the superconducting gap of the electron system at the LaAlO₃-SrTiO₃ interface using tunneling spectroscopy. The features are found only in a number of samples and depend upon the thermal-cycling history of the samples. The features consist of a peak at zero energy and other peaks at finite energies, symmetrically placed around zero energy. These peaks disappear, together with the superconducting gap, with increasing temperature and magnetic field. We discuss the likelihood of various physical mechanisms that are known to cause in-gap features in superconductors and conclude that none of these mechanisms can easily explain the results. The conceivable scenarios are the formation of Majorana bound states, Andreev bound states, or the presence of an odd-frequency spin triplet component in the superconducting order parameter [1].



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Electron Accumulation and Emergent Magnetism at the $\text{LaMnO}_3/\text{SrTiO}_3$ Heterointerface

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Emergent phenomena at polar-nonpolar oxide interfaces have been studied intensely in pursuit of next-generation oxide electronics and spintronics. Despite these studies, the production of higher-order effects (beyond conduction) and control of the ultimate size scaling of such emergent phenomena have been limited. In this presentation, we report the disentanglement of critical thicknesses for electron reconstruction and the emergence of ferromagnetism in polar-mismatched $\text{LaMnO}_3/\text{SrTiO}_3$ (001) heterostructures. Using a combination of element-specific X-ray absorption spectroscopy and dichroism, and first-principles calculations, interfacial electron accumulation and ferromagnetism have been observed *within* the polar, antiferromagnetic insulator LaMnO_3 . Our results show that the critical thickness for the onset of electron accumulation is as thin as 2 unit cells (UC), significantly thinner than the observed critical thickness for ferromagnetism of 5 UC. The absence of ferromagnetism below 5 UC is likely induced by the electron over-accumulation. In turn, by controlling the doping of the LaMnO_3 , we are able to neutralize the excessive electrons from the polar mismatch in ultrathin LaMnO_3 films and thus enable ferromagnetism in films as thin as 3 UC, extending the limits of our ability to synthesize and tailor emergent phenomena at interfaces and demonstrating manipulation of the electronic and magnetic structures of materials at the shortest length scales.

Room-temperature Blue Luminescence from Highly Electron-doped TiO_x Nanostructures on the Surface of SrTiO_{3-δ} (001)

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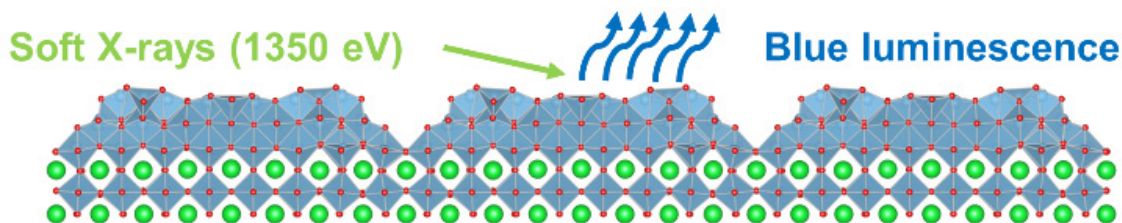
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Tuning the defect structure in the near-surface and interface regions of complex oxides is critical in achieving the desired electronic and optical properties. Doping electrons into the perovskite oxide SrTiO₃ results in a myriad of fascinating properties, including superconductivity [1] and also the formation of a two-dimensional electron gas when interfaced with LaAlO₃ (001) [2]. One of the ways electron doping manifests itself in SrTiO₃ is the emission of blue light [3]. High electron doping can be stabilized on air-stable surfaces of SrTiO₃ (001) where the oxygen coordination of Ti differ significantly from the bulk, with potential electron doping up to 0.57 *e*⁻/square lattice parameter [4], which may be of use in interface-based oxide electronics. Here we report on the observation of a relatively strong contribution to the blue luminescence from TiO_x nanostructures formed on the surface of SrTiO_{3-δ} (001) with an initially TiO₂-terminated stepped-and-terraced surface after a DC-resistive heating process. By examining the dynamics of the blue luminescence, we quantify the doped electron concentrations in the bulk and surface regions, which reveals enhanced electron doping in the surface TiO_x nanostructure that explains the two-component blue luminescence behavior.



Atomic structure of the 'triline' TiO_x nanostructure on SrTiO₃ (001) obtained from Ref. [4] responsible for the surface contribution to the blue luminescence.

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Dielectric characteristics of $\text{La}_{0.5}\text{Cr}_{0.5}\text{TiO}_{3+\delta}$ and visible light modulation

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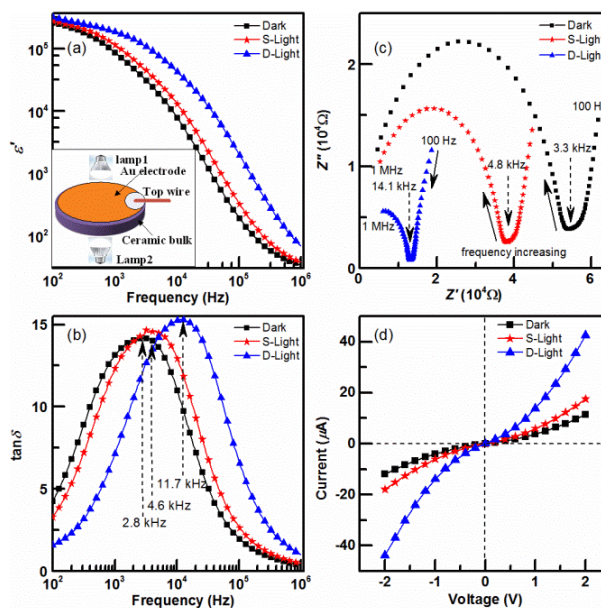
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$\text{La}_{0.5}\text{Cr}_{0.5}\text{TiO}_{3+\delta}$ ceramic sample is prepared via traditional solid-state reaction route. Frequency and temperature dependence of dielectric permittivity were studied in the range from 10^2 to 10^6 Hz and from 77 to 360 K, respectively. We observed that extraordinarily high low-frequency dielectric constants appear at room temperature and dielectric relaxation peaks shift to higher temperature with increasing frequency. In the dc-bias studies, we also found that the dielectric permittivity has obviously dc-bias dependence in low frequency, but independence as the frequency above 14 kHz. Interestingly, the dielectric characteristics of the sample have obvious light dependence at room temperature within the measured frequency range. The results have demonstrated that visible light improves the dielectric properties of the ceramics by means of I - V and complex impedance analysis.

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The dielectric and I - V characteristics of the $\text{La}_{0.5}\text{Cr}_{0.5}\text{TiO}_{3+\delta}$ sample under visible light conditions, (a) the dielectric constant ϵ' , (b) the dielectric loss tangent $\tan\delta$, (c) the cole-cole plot, and (d) the I - V characteristics of the sample on room temperature under different visible light conditions. The inset in (a) is a schematic of dual visible light sources on sample. Dark: without light, S-Light: single visible light source on side of sample, D-Light: dual visible light sources on both sides of sample, respectively.

SmNiO₃/NdNiO₃ superlattices

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Among the numerous families of perovskite oxides, the rare earth nickelates (chemical formula RNiO₃, R = rare earth) stand out for their unique metal to insulator transition (MIT), accompanied by a symmetry lowering from an orthorhombic to a monoclinic structure and a breathing distortion of the NiO₆ octahedral units [1-3]. Moreover, they display an unusual antiferromagnetic (AF) ordering at $T_{\text{Néel}} \leq T_{\text{MI}}$. Notably, the critical temperature for the MIT (T_{MI}) is a function of the Ni-O-Ni bond angle characterizing each member of the family, which can be tuned, for example, by substitution of the A-cation or epitaxial strain [4-6]. Here, we adopt a further strategy to inspect the evolution of structural distortions in RNiO₃-based heterostructures and their impact on the electronic properties of the system. We fabricated $[N(\text{SmNiO}_3)/M(\text{NdNiO}_3)]_n$ superlattices by off axis magnetron sputtering, where M , N are the number of unit cells of, respectively, the SmNiO₃ (SNO) and NdNiO₃ (NNO) layers and n is the number of repetitions. In bulk, SNO displays $T_{\text{MI}} > T_{\text{Néel}}$ and NNO shows $T_{\text{MI}} = T_{\text{Néel}}$. Electrical transport measurements on these heterostructures show MITs which are tuned by the periodicity of the superlattices.

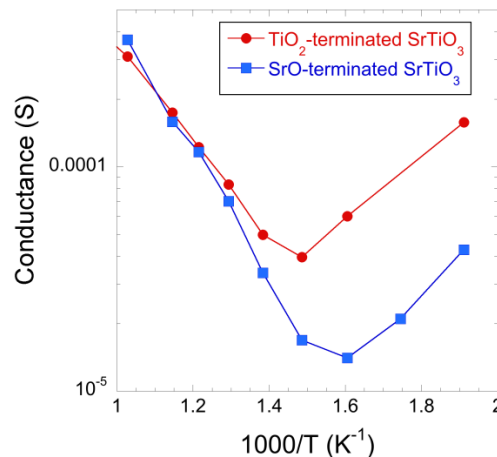
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Interfacial Electrical Conductance Behavior in Thin Film Epitaxial (001) In_2O_3 / SrTiO_3 Heterostructures

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In this presentation, the elevated temperature electrical conduction behavior of heterostructures consisting of In_2O_3 thin films grown epitaxially on (001)-oriented SrTiO_3 single crystal substrates will be described. At temperatures below approximately 500°C , a metal-like temperature dependence is observed, while at higher temperatures the semiconducting behavior expected for In_2O_3 is seen. The (001) In_2O_3 / SrTiO_3 interface is polar, and the behavior is qualitatively similar to that reported for LaAlO_3 / SrTiO_3 heterostructures [1], another polar oxide system. In both cases, the temperature dependence of the conduction behavior can be attributed to parallel contributions from different regions of the sample, including a metal-like heterointerface region that dominates at lower temperatures. However, in the present case the interface is between an n-type semiconductor (In_2O_3) and a bulk insulator (SrTiO_3), rather than between two insulators. Interestingly, we find that the magnitude of the metal-like component of the conductance depends significantly on whether the In_2O_3 film has been grown on a TiO_2 - or SrO -terminated SrTiO_3 surface, indicating that the detailed nature of the heterointerface plays a strong role in controlling the observed conduction behavior.



Comparison of the temperature-dependent conductance of heterostructures consisting of equal thickness (13.5 nm) In_2O_3 films grown on TiO_2 - or SrO -terminated (001) SrTiO_3 . Both samples were equilibrated at each temperature in a 150 Torr oxygen partial pressure environment.

In_2O_3 films were grown at 700°C by RF-magnetron sputtering on SrTiO_3 substrates with controlled TiO_2 - or SrO -termination, prepared following published recipes [2]. Four-point conduction measurements in an environmental probe station were obtained while monitoring sample equilibration under controlled temperature and oxygen partial pressure conditions. Observations of the effects of oxygen partial pressure on conduction revealed possible defect mechanisms relevant to the observed behavior that will also be discussed. The work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

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Superconductivity at $\text{LaAlO}_3/\text{Sr}_{1-x}\text{Ca}_x\text{TiO}_3$ interfaces

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The superconducting state of doped SrTiO_3 is somehow unique: it appears at the lowest carrier concentration in a material close to a ferroelectric order.

Recent work has investigated the influence of ferroelectric fluctuations on superconductivity [1]. This theoretical study predicts an enhancement of the superconducting transition temperature (T_c) for samples in the vicinity of the ferroelectric quantum critical point (QCP). Experimental studies in ferroelectric $\text{SrTi}^{18}\text{O}_3$ [2] and $\text{Sr}_{1-x}\text{Ca}_x\text{TiO}_3$ ($0.002 < x < 0.02$) [3] have indeed revealed that T_c is higher for ^{18}O and Ca-doped SrTiO_3 samples.

In this study, we investigate superconductivity at $\text{LaAlO}_3/\text{Sr}_{1-x}\text{Ca}_x\text{TiO}_3$ interfaces to clarify how the 2D electron liquid at the interface is affected by the ferroelectric QCP. A clear metallic behavior is observed in SrTiO_3 (6 u.c.)/ LaAlO_3 (5 u.c.)/ $\text{Sr}_{1-x}\text{Ca}_x\text{TiO}_3$ ($x = 0.005$ and 0.01) heterostructures, with sheet resistance values similar to the ones of the $\text{LaAlO}_3/\text{SrTiO}_3$ system. We will discuss the superconducting properties of the $\text{LaAlO}_3/\text{Sr}_{1-x}\text{Ca}_x\text{TiO}_3$ interfacial system under field effect.

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Insulating magnetic oxides in spintronics. The key role of interfaces

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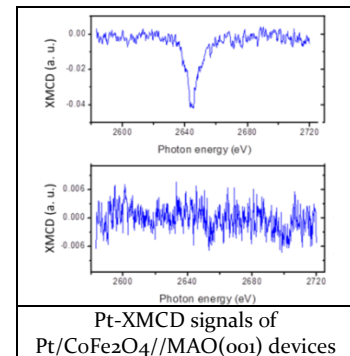
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Pure spin currents are seen as an alternative for lower energy consumption spintronics. It involves the manipulation of spin currents in absence of charge currents, thus getting rid of Joule dissipation. In a particular realization, a ferromagnetic insulating oxide layer (FMI) is covered by a non-magnetic metallic layer (NM) forming NM/FMI stacks. Current-induced spin flow in the NM layer is reflected back at the FMI interface or it diffuses across (denoted “spin mixing conductance G ”), depending on the magnetization of FMI at the interface. An anisotropy of G has been observed in Pt/CoFe₂O₄ bilayers grown along different crystallographic directions [1]. More intriguing is the observation of a distinct magnetic field dependence of the so-called spin magnetoresistance (SMR), related to G , that of the FMI film magnetization [2]. This finding was used to claim that SMR is a sensitive probe to magnetization at the buried NM/FMI interface.

Here, we address some of these pivotal questions. We focus on two devices: a) Pt(4 nm)/CoFe₂O₄(10-60 nm)//SrTiO₃(111) and b) Pt(4 nm)/CoFe₂O₄(28 nm)//MgAl₂O₄(001). In the first case (a), we only changed the magnetic microstructure of the CoFe₂O₄ layer by changing its thickness. In the second case (b), we changed the temperature for sputtering the Pt layer. We characterized the magnetic response $M(H)$ by conventional magnetometry and measured the spin Hall magnetoresistance (SMR). We measured the X-ray absorption at Pt and (Co, Fe) edges and determined the XMCD. Sample characterization is completed by using STEM-EELS. Two main conclusions emerge. a) In Pt(2-6 nm)/CoFe₂O₄(10-60 nm)//SrTiO₃(111), the SMR signal nicely mimics the (Co,Fe) XMCD signals which differ from the $M(H)$. This indicates that surface magnetization of CFO is different from bulk and we obtain hints of the microstructural origin of the difference. We assess the absence of any magnetic moment in Pt, thus excluding proximity effects in the measured SMR signals [3] and b) A similar situation holds in Pt(4 nm)/CoFe₂O₄(28 nm)//MgAl₂O₄(001) if Pt is grown at room-temperature but induced magnetism at Pt is observed if grown at high temperature. These results show that interfaces play a crucial role in these novel devices and much work is required for proper control and understanding.



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Run-away tail (RAT) in SrTiO₃ accumulation layers

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We study in our work¹ the low temperature conductivity of the electron accumulation layer induced by the very strong electric field on the surface of the SrTiO₃ (STO) sample. Due to the strongly nonlinear lattice dielectric response, the three-dimensional density of electrons $n(x)$ in such a layer decays with the distance from the surface x very slowly as $n(x) \propto 1/x^{12/7}$. We show that when the mobility is limited by the surface scattering the contribution of such a tail to the conductivity diverges at large x because of growing time electrons need to reach the surface (see Fig. 1). We explore truncation of this divergence by the finite sample width, by the bulk scattering rate, or by the crossover to the bulk linear dielectric response with the dielectric constant κ . As a result we arrive at the anomalously large mobility, which depends not only on the rate of the surface scattering, but also on the physics of truncation. Similar anomalous behavior is found for the Hall factor, the magnetoresistance, and the thermopower as well. For the Hall factor, another truncation mechanism by the large magnetic field gives rise to the nonlinear Hall effect observed experimentally². The thermopower S depends on the thickness W as $S \propto W^{8/7}$, which is also consistent with experimental observations³ (see Fig. 2).

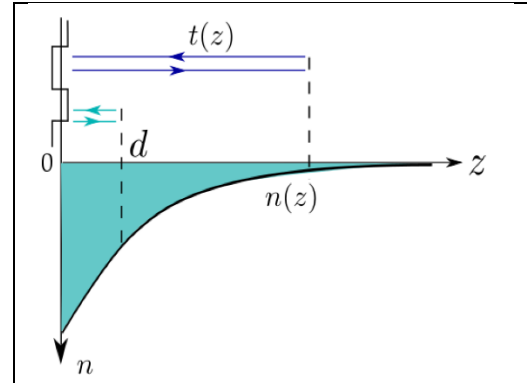


Fig. 1: Schematic plot of electron relaxation by surface roughness scattering. The ups and downs on the surface represent the roughness. Electrons in the tail have a much larger travel time to the surface and thus a much larger relaxation time than electrons in the main body of the distribution close to the surface.

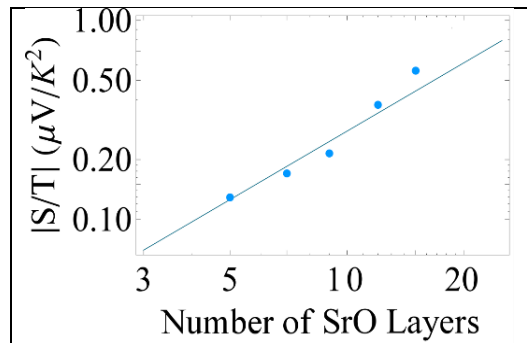


Fig. 2: Comparison of experimental data and RAT prediction for the absolute value of the thermopower S divided by the temperature T as a function of the STO sample width in the GTO/STO/GTO quantum wells represented by the number of STO layers.

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Applications of electron microscopy imaging and spectroscopy to understand structure-properties relationships in complex functional materials

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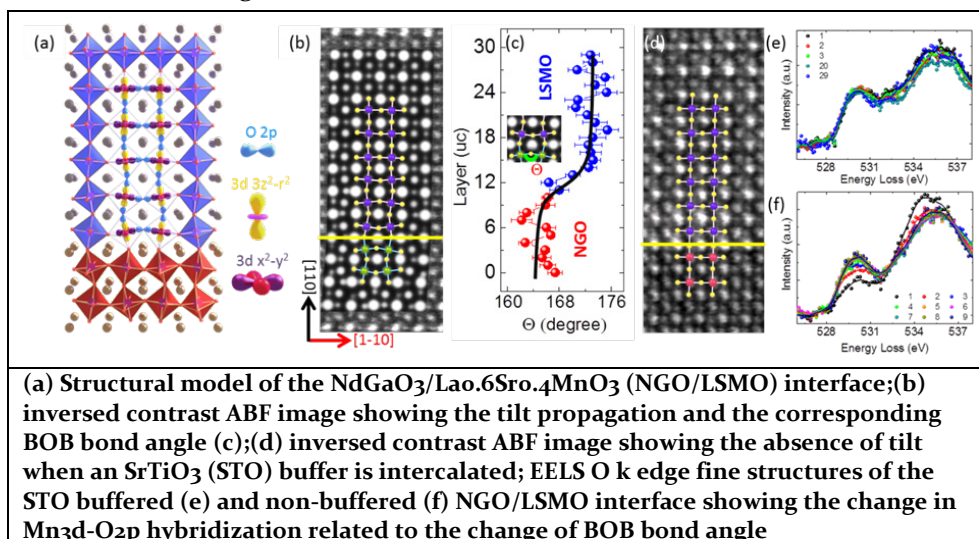
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The study of novel physical properties appearing when two materials are interfaced has become one of the major fields of research in solid-state physics over the last decade. As the materials involved in those new physical phenomena are often complex oxides, many factors (such as strain, oxygen stoichiometry, cation intermixing, electronic reconstructions) have to be considered when discussing their origin. If the two interfaced materials have different octahedral tilt systems, one has to adapt to the other. Electron microscopy imaging (HAADF, ABF) and EELS spectroscopy are key tools for their study.

I will be reporting recent results enlightening the effect of oxygen octahedral coupling on the change of the magnetic easy-axis in some ferromagnetic manganite films. [1] I will show how this change of magnetism can be related to a change in hybridization between the metal and the oxygen orbitals [2]. Another example of hybridization effects on physical properties will be presented in RNiO_3 ($\text{R}=\text{Sm}, \text{Nd}$) superlattices will be also presented.



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Competition of Orbital Polarization Sources in a Nickelate/Aluminate Superlattice

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A topic of recent interest in the study and design of complex oxide heterostructures has been the orbital engineering of the 3d orbital manifold, specifically by engineering the energy splitting of the e_g orbitals in order to create an orbitally polarized state. This should, in principle, make the band structure resemble that of cuprate superconductors. An important resulting question is: what are the causes of orbital polarization in a heterostructure?

In this work we explore the effect of three interacting causes through a combination of first principles (density functional theory) calculations and slave-boson approaches in a $\text{NdNiO}_3/\text{NdAlO}_3$ superlattice. We show how electrostatic effects due to the relative ionicity of the atoms in the two materials lead to local electrostatic effects within the Ni oxygen octahedra, leading to an energy splitting that leads to a previously undiscussed source of orbital polarization. We show how this effect, correlation-induced band narrowing and quantum confinement lead to the resulting orbital polarization in the Ni e_g manifold.

The research reported in the paper has been supported by the National Science Foundation via Grant MRSEC DMR 1119826.

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Effective Correlation Tuning via Mixed-Phase Pyrochlore Iridate Thin-Films

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Pyrochlore iridates ($R_2\text{Ir}_2\text{O}_7$, R: rare-earth lanthanides) have attracted substantial attention for predictions of topological properties emerging from frustrated magnetic spin structures (all-in-all-out AIAO ordering) in the presence of strong spin-orbit interaction and effective electron correlation (U_{eff}) [1]. A method for probing this multi-interaction phase space is by monotonically tuning U_{eff} , which is achieved experimentally by altering the pyrochlore iridate lattice size, i. e. U_{eff} increases as the size of R decreases. Although this picture is quite consistent with the transport of polycrystalline crystals [2], the Weyl phase has remained elusive.

In order to tune U_{eff} , we have synthesized a series of mixed phase pyrochlore iridate thin-films ($(A,B)\text{Ir}_2\text{O}_7$ for $(A,B) = (\text{PrNd})$; $(\text{Nd}_{1.5}\text{Sm}_{0.5})$; (NdSm) ; $(\text{Nd}_{0.5}\text{Sm}_{1.5})$) on the wide-band insulator Y:ZrO₂ (YSZ) using a combination of pulsed laser deposition and a high-temperature anneal in ambient. As the average rare earth ionic size is increased from Sm to Pr, the thin-film lattice parameters systematically increase, as revealed by x-ray diffraction. This consequently induces a monotonic decrease in the metal-to-insulator transition temperature and is consistent with reducing U_{eff} in these mixed-phase films (Fig. 1). We have also observed an interesting crossover in rotational magnetotransport when the applied field H is parallel to the $[110]$ film direction, where a sharp conductivity spike observed in $\text{PrNdIr}_2\text{O}_7$ is replaced by a sharp conductivity drop for the remaining films with smaller lattice sizes. We speculate that in this field configuration, H flips the spins of the rare earth sites, which act to generate a maximal number of domain walls. The smaller U_{eff} in $\text{PrNdIr}_2\text{O}_7$ allows for conducting domain walls whereas the larger U_{eff} in the remaining samples induces insulating domain walls, which explains the contrasting conductance behavior. The mixed-phase pyrochlore iridate thin-films allow for position-dependent measurements not accessible by polycrystalline studies and offer an effective means for tuning U_{eff} across distinct electronic phase boundaries.

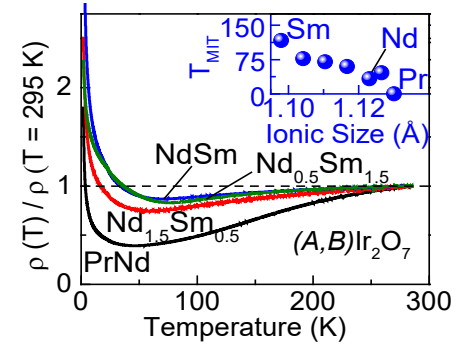


Figure 1. Metal-insulator transitions (MIT) in electric transport of $(A,B)\text{Ir}_2\text{O}_7$ / YSZ (111). All films undergo an MIT which sequentially increases in temperature as the average rare-earth ionic radius is decreased (inset).

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Solution-processed metal-oxide semiconductors for neuromorphic electronics

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Superb electrical characteristics of amorphous metal-oxide semiconductors (AOS) have been shown to be promising for next-generation electronics which require low-cost fabrication and low-power consumption [1]. Recently, low temperature and large-area processes for solution-processed AOS compatible with flexible substrates have been of technical interest in neuromorphic electronics [2]. However, such AOS still exhibits relatively poor material and electronic properties compared to that achieved through vacuum-process fabrication because of the unexpected presence of charge impurities, including oxygen and water molecules, as well as residues of the sol-gel process in uncontrolled environments [3].

In this presentation, we will demonstrate high-quality solution-processed AOS for neuromorphic electronics. By employing a structural integration of metal-oxide semiconducting films and an optimization for fabrication process, all the key metrics of solution-processed AOS were improved. We will also investigate the stability mechanism of solution-processed AOS by analyzing the binding energy and the impurity scattering. We note that this work can open up a practical approach to improve the material and electronic characteristics of solution-processed AOS for neuromorphic electronics.

Acknowledgment:

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Synchrotron operando measurements: Combined X-ray absorption spectroscopy and magnetotransport measurements with magnetic tunnel junctions

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In X-ray absorption spectroscopy (XAS), the energy of soft X-ray photons is tuned so as to drive core level excitations of a specific atomic species within the sample. The resulting absorption spectrum yields information on the quantity and charge state of these atoms present, on their chemical environment and their resulting electronic/magnetic properties.

The principle of operando studies is to use the capabilities of X-rays to probe some properties of a sample under external stimulus, applied in situ. While usually one has to separate the materials science and device studies, operando measurements offer the possibility to probe the devices' properties in real operating conditions and thus to establish causal link between material properties and device operation.

In this work^{1,2}, we combine X-ray absorption spectroscopy (XAS) and magnetotransport measurements to study CoFeB/MgO/CoFeB magnetic tunnel junctions. Specifically, we measured the devices electrical current, which involves charge transport across different atomic species and interfaces, while these atoms absorb soft X-rays. X-ray absorption is found to affect magnetotransport when the photon energy and linear polarization are tuned to excite Fe-O bonds parallel to the MTJ's interfaces. This explicit link between the device's spintronic performance and these Fe-O bonds, although predicted, challenges conventional wisdom on their detrimental spintronic impact.

The technique opens interdisciplinary possibilities to directly probe the role of different atomic species on device operation, and could considerably simplify the materials science iterations within device research.

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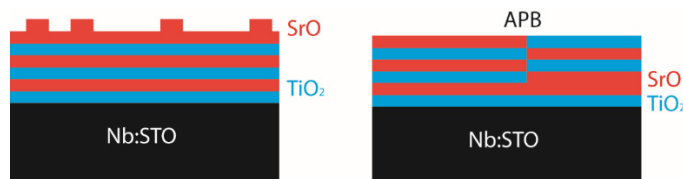
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Tailoring the switching performance of resistive switching SrTiO₃ devices by SrO interlayer engineering

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A central challenge in today's scientific community is the need for faster and smaller non-volatile memory devices. A potential candidate for achieving such devices is the mechanism of resistive switching. SrTiO₃ (STO) is a model material for filamentary resistive switching oxides. This is due to its well-known defect chemistry and the intense research on its switching behavior. Nevertheless there are still plenty of open questions regarding the in depth understanding of resistively switching STO.

In the past we have shown that epitaxially grown STO with an excess of Sr shows an increased memory window and an improved retention of the low resistive state¹. In order to understand the improved switching behavior of Sr-rich STO thin film devices, we investigated the possibilities of Sr-excess compensation during pulsed laser deposition growth. We considered two extreme cases of Sr-excess accommodation. Namely the formation of stacking faults within the thin film and the segregation of SrO on top of the thin film (see figure). We were able to artificially engineer these two cases by intentionally depositing additional SrO at the respective position. We observed that thin film stacks with SrO at the bottom interface results in forming free devices, which might be attributed to the formation of extended defects such as antiphase boundaries² which act as preformed filaments. On the other hand introducing additional SrO to the top interface results in an improved memory window and retention. By controlling the amount of SrO on top we were even able to increase the memory window and improve the retention of the low resistive state. We attribute the improved retention of the low resistive state to the impeded reoxidation of the conducting filament by the SrO layer³. These results provide a pathway to a rational design of resistive switching devices with the possibility of prepositioning switching filaments.



Possibilities for SrO excess accommodation during epitaxial growth of STO. Left: The outgrowth of additional SrO on top of the thin film. Right: Formation of antiphase boundaries as a result of stacking faults.

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In-situ X-ray Studies of $\text{In}_2\text{O}_3\text{:CeO}_2$ Thin Films and Nanostructures

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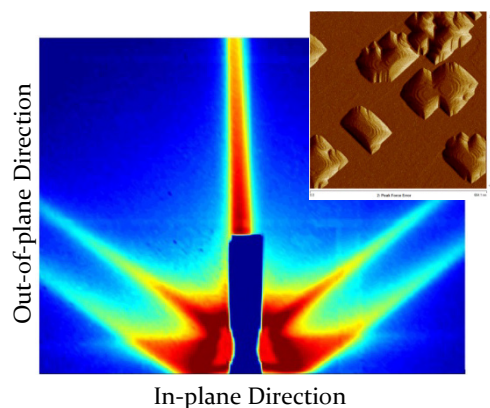
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One of the most promising methods for manipulating the properties of oxide materials is through the formation of phase separated nanocomposites. In nanocomposite materials the crystal lattices of two or more distinct phases interact to influence the stability and properties of one another. This complex interaction between materials has been studied as a method for the formation of multi-ferroics, fast ionic conductors, and optical meta-materials [1-3]. We have utilized in-situ x-ray scattering to studies the processes that govern the formation of oxide nanocomposite.

We will present studies on the formation of $\text{In}_2\text{O}_3\text{:CeO}_2$ alloys and two phase mixtures with an emphasis on understanding the processes that dictate when a phase separated nanocomposite will form and the microstructure it will adopt. In_2O_3 and CeO_2 are well suited for the study of nanocomposite formation due in part to their range of interesting electrical, optical and ionic properties [4-6]. We will discuss how extrinsic parameters in the synthesis process and intrinsic properties of the material system can be used to guide the nanocomposite formation process. These results suggest new strategies for controlling nanocomposite formation both in this material system and oxides in general.

This work is supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences.



In-situ GISAXS and ex-situ AFM of an $\text{In}_2\text{O}_3\text{:CeO}_2$ nanocomposite film.

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Epitaxial growth of high quality SrFeO_3 films on (001) oriented $(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{TaAlO}_6)_{0.7}$

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Growth of strontium ferrite SrFeO_3 with stoichiometry of (1:1:3) is always an issue due to the large negative charge transfer energy of Fe^{4+} . According to previous reports, a very high oxygen pressure (several hundred bar) annealing procedure is always applied for fully stoichiometry. Here, we report a layer by layer growth of SrFeO_3 on (001) oriented $(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{TaAlO}_6)_{0.7}$ using ozone assisted molecular beam epitaxy. According to the x-ray diffraction, our film is epitaxial with the out-plane lattice constant $\sim 3.83 \text{ \AA}$, corresponding to the small radius of Fe^{4+} . Upon cooling from room temperature, the film's resistivity decreased from $750 \mu\Omega \cdot \text{cm}$ to $150 \mu\Omega \cdot \text{cm}$ with two identifiable transition points near 110 K and 60 K. The resistivity is hysteretic between cooling and warming through the 60 K transition. As far as we know, all these behaviors have been observed in SFO bulks only before this work. We further annealed our samples under various conditions, resistivity changes a lot, especially at low temperatures, along with the crystal lattice parameter c . We have identified conditions under which this change is reversible. We attribute changes in resistivity and out of plane lattice parameter to the movement of oxygen ions in Fe octahedra. Our results are meaningful for the further fundamental research in SFO and applications in electronic devices and energy related solid oxide fuel cell.

Vertical Transport in Epitaxial BaTiO₃-Germanium Heterostructures

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Recent advances in epitaxial growth of BaTiO₃ thin films on semiconductors open new opportunities for coupling ferroelectric complex oxides to conventional complementary metal-oxide-semiconductor (CMOS) devices. Understanding the conduction mechanism in BaTiO₃-semiconductor heterostructures is a key step to developing CMOS-compatible non-volatile ferroelectric devices based on BaTiO₃. Here we investigate vertical transport in epitaxial BaTiO₃-Ge heterostructures. BaTiO₃ is grown on p-type Ge via molecular beam epitaxy (MBE) followed by electron-beam lithography to define device structures. *In-situ* reflection high-energy electron diffraction (RHEED) and *ex-situ* X-ray diffraction (XRD) confirm that the BaTiO₃ is single crystalline. Transport characterization is carried out from room temperature to 2 Kelvin. We find the conduction at low temperature is dominated by tunneling at negative gate bias, thus enabling tunnel transport devices.

Tuning magnetic anisotropy by interfacially engineering the oxygen coordination environment in a transition-metal oxide

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Metal-oxygen bonds in transition-metal oxides are responsible for a broad spectrum of functional properties, and atomic-level control of the bonds is a key for developing future oxide-based electronics. Artificial heterostructures with chemically abrupt interfaces consisting of dissimilar oxides have provided a good platform for engineering novel bonding geometries that could lead to emergent phenomena not seen in bulk oxides. Here we show that the Ru-O bonds (or oxygen coordination environments) of a perovskite, SrRuO₃, can be controlled by heterostructuring SrRuO₃ with a thin (0–4 monolayers thick) Ca_{0.5}Sr_{0.5}TiO₃ layer grown on a GdScO₃ substrate (1,2). We found that a Ru-O-Ti bond angle characterizing the SrRuO₃/Ca_{0.5}Sr_{0.5}TiO₃ interface structure can be engineered by layer-by-layer control of the Ca_{0.5}Sr_{0.5}TiO₃ layer thickness, and that the engineered Ru-O-Ti bond angle not only stabilizes a Ru-O-Ru bond angle never seen in bulk SrRuO₃ but also tunes the magnetic anisotropy in the entire SrRuO₃ layer. We also show that an orbital magnetic moment plays a key role in tuning the magnetic anisotropy through the metal-oxygen bond (3). The results demonstrate that interface engineering of the metal-oxygen bonds is a good way to control additional degrees of freedom in functional oxide heterostructures.

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Spin-orbit coupling and electronic correlations in Hund's metal : Sr_2RuO_4

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We investigate spin-orbit coupling (SOC) effects on the correlated electronic structure of Sr_2RuO_4 using the dynamical mean-field theory. The approach reproduces experimental results on the electronic structure of Sr_2RuO_4 , resolving issues on strong correlations and SOC effects with remarkable consistency. It is shown that SOC does not affect the nature of electronic correlation driven by Hund's coupling. The renormalization of bandwidths has strong energy dependence which is a signature of Hund's metal. On one hand, electronic correlation enhances effective SOC constants without subtle energy dependence, reflecting atomic nature of SOC in the presence of electronic correlation. These aspects are essential to describe experimental results, (i) SOC driven degeneracy lifting at Γ point, (ii) SOC effective Fermi surface, and (iii) correct renormalization of bands in which SOC is not effective. The result opens a new avenue on the SOC effective strongly correlated Hund's metal.

Tuning Magnetic, Structural and Electronic Interactions at Polar Manganite Interfaces

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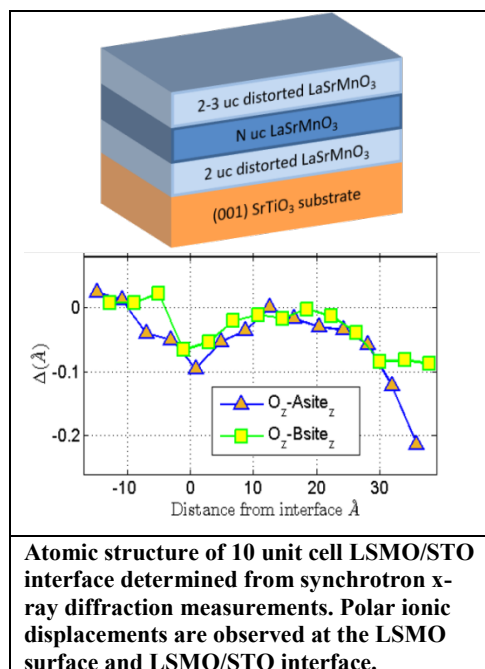
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Chemical, orbital, electronic and structural interactions at polar oxide interfaces and surfaces have led to the realization of novel physical properties including high mobility two-dimensional electron gases and interfacial magnetism. These interactions also provide a route to manipulate the electronic and magnetic properties of complex oxide thin films.

Using high-resolution synchrotron diffraction imaging, we investigate the structural interactions at the interface between the polar half-metallic ferromagnet $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) and non-polar SrTiO_3 (STO). We observe polar rumplings extending 2-3 unit cells away from the LSMO film surface and LSMO/STO interface in response to the surface and interfacial polar discontinuities. [1]

By monitoring the evolution of these structural distortions as a function of interfacial and surface composition achieved by tuning the La/Sr ratio at the film surface and interface, a strong link is established between polarity-driven atomic reconstructions and the observed magnetic and electronic dead-layer effects observed in the rare-earth manganites. These results demonstrate the efficacy of tuning interfacial polar discontinuities as a route to engineer the functional properties of two-dimensional complex oxide materials.



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Large enhancement of ionic conduction in vertically aligned single crystalline oxide nanosuperlattices

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The development of solid-state ionic electrolytes with high ionic conductivity is strongly required to develop next generation energy and environmental technologies, such as solid oxide fuel cells (SOFCs) and oxygen separation membranes. Over the last few decades, two dimensional (2D) oxide nanostructures, in particular, oxide multilayer heterostructures have brought tremendous advances in enhancing the ionic conductivity of oxides. However, new design concepts are required to satisfy the technological needs such as thickness in micron-scale and fast ion transport along the out-of-plane direction for realization of the commercial applications. One dimensional (1D) oxide nanostructures have shown its feasibility as a new type of ionic electrolytes due to large length-to-width aspect ratio, highly crystalline faceted structure, and high surface-to-volume ratio. However, a lack of understanding on the growth control of 1D oxide nanostructures at the atomic scale impedes implementation of low dimensional oxide materials for advanced energy and environmental devices. Here, we report a completely new form of oxide nanostructures for achieving superior out-of-plane ionic conductivity by a combination of one dimensional oxide system¹ and multilayer oxide system using properly balancing the kinetic and thermodynamic nonequilibrium pulsed laser epitaxy processes. We synthesized vertically aligned micron-thick oxide superlattices composed of CeO₂ and Y₂O₃ with high surface area and high porosity. The ionic conductivity of oxide nanosuperlattices was enhanced by up to four orders of magnitude compared to undoped CeO₂ through electrochemical impedance spectroscopy. Scanning transmission electron microscopy was utilized to investigate the structural feature of oxide nanosuperlattices and revealed an intriguing space charge layer formation due to ionic reconstruction at CeO₂-Y₂O₃ interfaces. In this presentation, the underlying mechanism of the growth of such unique oxide nanosuperlattices and the origin of the enhanced ionic conductivity will be discussed. Our finding provides deeper insight into an enhancement of oxygen ionic conductivity and suggests the possibility of using a new form of oxide nanostructures in advanced energy and environmental applications.

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Spin States Control in LaCoO₃-based Heterostructures

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By modifying the electronic orbital configuration, we can access multiple competing spin-states of cobalt, including low-spin, intermediate-spin, and high-spin states, due to the balance between the crystal field, magnetic exchange, and Hund's coupling. Careful design of LaCoO₃-based heterostructures allows one to control the spin states of Co 3d orbitals and thus manipulate their macroscopic properties through charge transfer, interfacial coupling and dimensional confinement.

In the model system (LaCoO₃)_m/(LaTiO₃)_n, we tune the magnetism via interfacial electron transfer from Ti to Co. The charge transferred to Co changes the Co 3d valence state, allowing potential changes in Co spin state. The ferromagnetic order present in LaCoO₃ thin film is suppressed upon heterostructuring. In order to understand the effect of dimensional confinement and how the structural and electronic degrees of freedom are coupled, we also study (LaCoO₃)_m/(SrTiO₃)_m heterostructures. By comparing different mechanisms of confinement and charge transfer, we identify the spin and magnetic configurations and tune their macroscopic properties.

Oxygen defect engineering triggered by temperature cycling in $\text{La}_{0.7}\text{Sr}_{0.3}\text{CoO}_3$ film

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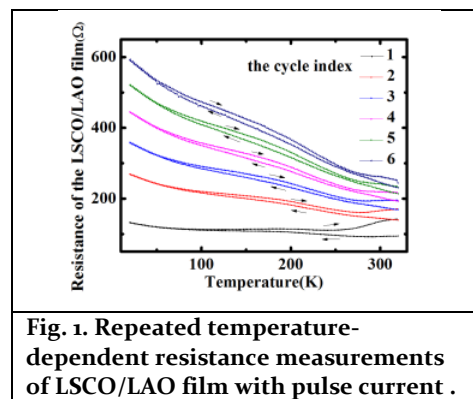


Fig. 1. Repeated temperature-dependent resistance measurements of LSCO/LAO film with pulse current .

It is expected to create novel materials with electronic, magnetic and structural properties by engineering oxygen defect in transition-metal oxides and their interfaces.[1-2]

The oxygen vacancy concentration can affect the redox activity of the $\text{La}_{0.7}\text{Sr}_{0.3}\text{CoO}_3$ (LSCO). However, the harsh reaction conditions at elevated temperatures and reducing atmosphere make the oxygen vacancies engineering difficult. Here, we report a new avenue to engineer the oxygen vacancies in LSCO film by temperature cycling assisted with pulse electric current. We deposited LSCO film on (001)-cut LaAlO_3 (LAO) and $(\text{LaAlO}_3)_{0.3}(\text{SrAl}_{0.5}\text{TaO}_3)_{0.7}$ (LSAT) substrates by pulsed laser deposition. The LSCO/LAO and LSCO/LSAT films undergo in-plane compressive strain and tensile strain, respectively. The resistance with the temperature cycling for the LSCO/LAO film was shown in Fig.1, where the numbers represent the cycle index of the transport measurements with temperature cycling. With the increase of temperature, the resistance in the heating process gradually increases and deviates from the one in the cooling process in each cycle, especially in the high temperature section ($>250\text{K}$). Moreover, the resistance in the succeeding cooling process is much higher than the one in both heating and cooling ones in the previous cycle. Similar phenomena were also observed in LSCO/LSAT film. In the oxygen-poor LSCO films, there are many O^{2-} ions moved to the electrical contact and released from the film, which were forced by the pulse current. In addition, the pulse current can slightly heat the electrical channels due to possible Joule heating effect, engineering the oxygen vacancies around the electrical channel. As a result, the Co^{4+} state in the film will transform to the Co^{3+} state by decreasing O^{2-} ions, leading to the increase of the resistivity. Furthermore, the enhanced O^{2-} ion mobility and larger lattice parameter make the oxygen vacancies form more easily in the high temperature range, thus resulting in a sudden increase in the resistivity approaching room temperature (as shown in Fig.1). Our work demonstrates a novel and highly-efficient method to engineer oxygen vacancies in the perovskite-type oxides and brings new opportunities in designing high-efficiency oxidation catalysts.

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Orbital control by interfacial oxygen engineering in nickelate heterostructures

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Epitaxial synthesis offers tremendous opportunities to artificially design materials with unique properties. New lattice structure, which differs completely from bulk, can be stabilized through geometric design. In this work, through the interfacial connection of dissimilar structures, we demonstrate a possibility of oxygen sublattice control in LaNiO_3 thin films by interfacing with the infinite layer SrCuO_2 . SrCuO_2 is known to form an infinite layer structure when the thickness is above 5 unit cells, whereas it forms a chain-type structure with the CuO_2 plane oriented along the normal to the interface. This thickness controlled oxygen sublattice manipulation at the interface of a LaNiO_3 - SrCuO_2 heterostructure is investigated by synthesizing $(\text{LaNiO}_3)_1/(\text{SrCuO}_2)_n$ superlattices ($n = 3-10$). Interestingly, we found an insulator to metal transition of $(\text{LaNiO}_3)_1/(\text{SrCuO}_2)_n$ superlattices when decreasing the SrCuO_2 thickness. X-ray spectroscopy revealed a different electronic structure of Ni in $(\text{LaNiO}_3)_1/(\text{SrCuO}_2)_n$ superlattices than that in bulk LaNiO_3 . Such an interfacial control of the nickelate's oxygen coordination opens a new route to engineering nickelate's electronic structure and orbital configuration, ultimately expediting the search for cuprate-like electronic structure in nickelate-based heterostructures.

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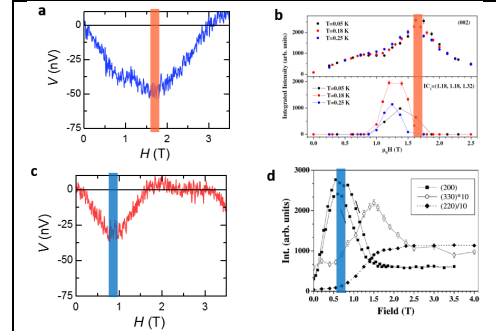
Probing short-ranged magnetic order in a geometrically frustrated magnet by spin Seebeck effect

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Geometrically frustrated magnets are a class of material in which a unique spin configuration cannot be established because of geometric constraints, which often leads to a rich phase diagram in these materials. Understanding the spin dynamics in these systems can lead to discoveries of new state of matter, such as quantum spin liquids. Currently, experimental methods for studying the complex magnetic phases have been mainly based on bulk magnetometries and neutron scatterings which are sensitive to only (relatively) long-ranged magnetic orders. Here, we demonstrate that by using spin Seebeck effect short-ranged magnetic orders in the geometrically frustrated hyperkagome antiferromagnet gadolinium gallium garnet (GGG) are detected at temperatures much higher than the ordering temperature of GGG. We find a quantitative agreement in terms of the field induced antiferromagnetic (AFM) order between the spin Seebeck responses and existing neutron scattering data. The reduction of SSE signal in response to these AFM order suggests that the flat band feature in frustrated magnets can be detected by SSE. In addition, we observe considerable modulations in spin Seebeck signal at high magnetic fields up to 9 Tesla, which points to unexplored physics in this model frustrated system.



Comparison between spin Seebeck signal and neutron scattering data. (a) and (c) Spin Seebeck signals measured with field applied along [110] and [100] GGG crystal axes, respectively. In both cases, a linear magnetic field dependent background has been removed. (b) and (d) Magnetic field dependence of the [002] AFM short range order in GGG probed by neutron scattering with field applied along [110] and [100] directions, respectively.

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Eliminating Surface Steps to Create Flat Heteroepitaxial Interfaces between Oxides and III-V Semiconductors

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Epitaxial integration of complex oxides with wide band gap polar semiconductors such as GaN (0002) presents the possibility for high-power, high-frequency, and high temperature GaN electronics by virtue of 2-D charge carriers at polar interfaces and possible access to non-linear dielectric properties. This work describes synthesis and characterization of such thin film heterostructures.

First, we will show how laser ablation provides pathways to lattice match epitaxial oxides that grow pseudomorphic to GaN. However, an ideal lattice match and a layer-by-layer growth mode, is not sufficient to create a globally ideal interface that can support a high mobility 2DEG. Given the symmetry difference between cubic oxides and wurtzite nitrides, GaN steps will always lead to stacking faults in the oxide overlayers, from which large defective grains evolve with thickness. Clearly, innovations from the nitride side are also needed.

To overcome this challenge we demonstrate how the combination of low defect density native GaN substrates and confined area epitaxy can completely eliminate surface steps over hundreds of microns creating macroscopic areas with only a single atomic terrace. Together, pseudomorphic oxide growth and flat GaN enable one to overcome the final obstacle to marrying cubic oxides with nitride semiconductors and to create interfaces that are truly semiconductor-grade.

We will show preliminary transport data for interface between (Mg_{0.5}Ca_{0.5})O and flat GaN with a uniform and step-free surface. While mobility values remain low, there is evidence for an interfacial population of charge carriers.

Flexoelectric and Electronic Structure Studies of Lanthanide Scandates

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Although first proposed in 1964 [1] and first observed in 1968 [2], flexoelectricity, the electromechanical coupling between polarization and strain gradient, in crystalline solids has remained largely unexplored. However, a number of experiments in the early 2000s helped to establish the flexoelectric effect as an important phenomenon in a large number of complex oxides, especially at the nanoscale [3]. Since applying a strain gradient is inherently symmetry breaking, flexoelectricity can be found in both centrosymmetric and non-centrosymmetric materials. This has generated interest in utilizing flexoelectricity in applications that have traditionally relied upon piezoelectricity and a number of demonstrations have already shown the feasibility of flexoelectric devices [4].

We report the existence of large flexoelectric responses in bulk lanthanide scandates measured with a three-point bending method [5]. The magnitude of the flexoelectric coefficient for [110] oriented DyScO₃ was measured to be 8.4 ± 0.4 nC/m. Although this coefficient is of the same order of magnitude as the flexoelectric coefficient in SrTiO₃, the dielectric constant of DyScO₃ at room temperature is an order of magnitude smaller than that of SrTiO₃. Therefore, the flexocoupling voltage (ratio of the flexoelectric coefficient to the dielectric constant) is an order of magnitude larger in DyScO₃ than in SrTiO₃ at 42 ± 2 V. This is large compared to typical flexocoupling voltages of 1-10 V [3], defying conventional flexoelectric theory which posits that flexoelectricity scales with dielectric constant and begs to question: what is the cause of the large flexoelectric response in DyScO₃? Results of temperature dependent x-ray photoelectron spectroscopy experiments in conjunction with density functional theory calculations which help elucidate the role of oxygen vacancies and 4f electrons in lanthanide scandates will also be presented.

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Electric field induced surface conduction in ferroelectric PbTiO_3

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Control of carrier density is an important technique to tune physical properties of functional oxide materials. While this technique is applied to many oxide materials such as semiconductors and Mott insulators, carrier doping into ferroelectric materials has not been explored much because it is believed that the mobile electrons in ferroelectric materials screen the polarization and suppress ferroelectricity. However, interaction between mobile carriers and ferroelectricity has attracted attention recently as it may induce fascinating physical properties such as superconductivity [1] or surface accumulation in a ferroelectric single crystal [2].

Here, we have successfully induced a surface conductivity in ferroelectric PbTiO_3 . To achieve a large carrier concentration, we adopted electric double layer transistor (EDLT) configuration (Fig. 1) which uses ionic liquid as a gate insulator. PbTiO_3 was grown by pulsed laser deposition on $\text{La}_{0.18}\text{Sr}_{0.82}\text{Al}_{0.59}\text{Ta}_{0.41}\text{O}_3$ (LSAT) substrate. Figure 2 shows transfer characteristics of an EDLT fabricated on the PbTiO_3 thin film at 300K. It demonstrates typical n-type operation with a high on/off ratio over 10^5 . At 300K, we have obtained a high sheet carrier density $\sim 5.0 \times 10^{13} \text{ cm}^{-2}$ which is above the maximum value achieved in conventional metal-oxide-semiconductor FET ($\sim 1.0 \times 10^{13} \text{ cm}^{-2}$).

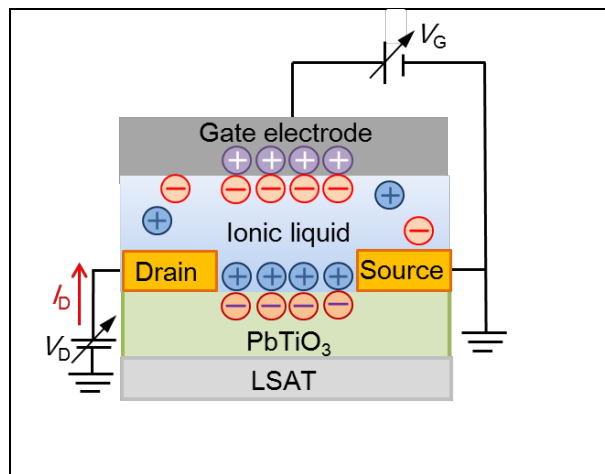


Figure 1 A schematic diagram of the EDLT configuration with a PbTiO_3 thin film channel.

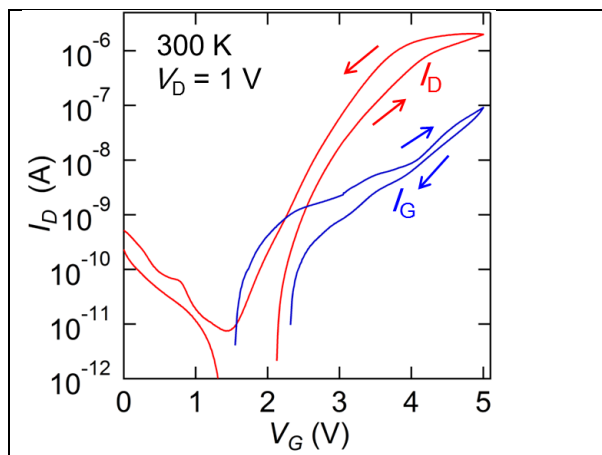


Figure 2 Transfer characteristics at 300K for the EDLT with a PbTiO_3 channel on a LSAT substrate.

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Spin-Hall Magnetoresistance in Canted and Collinear Magnetic Insulators

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The interplay between charge and spin currents at the interface between paramagnetic metals and magnetic insulators results in novel spintronic effects, such as the recently discovered spin-Hall magnetoresistance (SMR). It manifests itself in an unexpected dependence of the resistivity of a paramagnetic Pt layer (representing the most widely used spin-Hall material) on the magnetization of an adjacent ferromagnet, such as $\text{Y}_3\text{Fe}_5\text{O}_{12}$ or NiFe_2O_4 [1,2]. Its interpretation relies on the reflection or absorption of spin currents at the interface [1-3].

We first discuss our latest results on the proximity magnetism and the SMR effect in the Pt/ $\text{Y}_3\text{Fe}_5\text{O}_{12}$ system from a comprehensive investigation of different samples as a function of the thickness of the Pt layer and the stacking order of the Pt and $\text{Y}_3\text{Fe}_5\text{O}_{12}$ layers. We study the X-ray absorption spectra (XAS) and the X-ray magnetic circular dichroism (XMCD) at the Pt $L_{2,3}$ edges [4] and perform measurements of the angle-dependent magnetoresistance (ADMR) [1,2,5]. The results are fully consistent with the prediction of the SMR theory [4] and demonstrate that the SMR effect is governed by the net magnetization in the collinear ferrimagnet $\text{Y}_3\text{Fe}_5\text{O}_{12}$.

Second, in the canted ferrimagnet $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ the ADMR signal shifts by 90° at the magnetic compensation temperature [6]. Using atomistic spin simulations and XAS/XMCD experiments, we understand this observation in terms of the magnetic field and temperature dependent orientation of magnetic moments on different magnetic sublattices [6]. This enables an SMR-based investigation of non-collinear magnetic textures.

Third, in the antiferromagnetic insulators NiO and Fe_2O_3 we always observe this 90° shift of the ADMR signal at high magnetic fields. Since in the spin-flop phase the sublattice magnetizations are oriented perpendicular to the external magnetic field, this evidences that the SMR effect is governed by the sublattice magnetizations.

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Structure, Stoichiometry Effect and Electronic and Thermal Transport in MBE-Grown BaSnO₃ Films and Heterostructures

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We will present a comprehensive study of growth, structure, stoichiometry, and electronic and thermal transport properties of BaSnO₃ films grown via a novel radical-based hybrid molecular beam epitaxy (MBE) approach. High-resolution x-ray diffraction was used to confirm the growth of phase-pure, epitaxial BaSnO₃ film on SrTiO₃ (001) and LaAlO₃ (001) substrates with a bulk-like, unstrained lattice parameter of 4.116 Å. *In situ* reflection high-energy electron diffraction (RHEED), and scanning transmission electron microscopy (STEM) confirmed strain relaxation in BaSnO₃ occurs via forming misfit dislocations with a critical thickness of ~1 nm for BaSnO₃ (001)/SrTiO₃(001).

Films' cation stoichiometry was investigated using a combination of high-resolution XRD, Rutherford backscattering spectrometry (RBS), and thermal and electronic transport measurements demonstrating adsorption-controlled growth of BaSnO₃ accompanied by the "MBE growth window", i.e. a range of Sn/Ba flux ratios where only stoichiometric films grow.

Thermal conductivity value of ~ 13 W/mK - similar to that of the bulk single-crystal was achieved for stoichiometric films, decreasing to ~ 2 W/mK for Ba-rich films. An electron mobility of 120 cm²/Vs at 2×10²⁰-4×10²⁰ cm⁻³ was achieved for films grown on SrTiO₃(001). Irrespective of Ba- or Sn-rich films, carrier density and mobility decreased for non-stoichiometric films showing both Ba- and Sn-vacancies act as an acceptor-like defects in BaSnO₃.

Boltzmann transport modeling combined with T-dependent transport revealed RT electron mobility in doped BaSnO₃ films is limited largely by charged dislocation scattering and longitudinal optical phonon scattering. Finally, we will discuss metal-insulator transition, the role of ionized impurity scattering on mobility in BaSnO₃, and the pathways to modulation doping using heterostructure engineering. Funded in part by the NSF and the AFOSR YIP.

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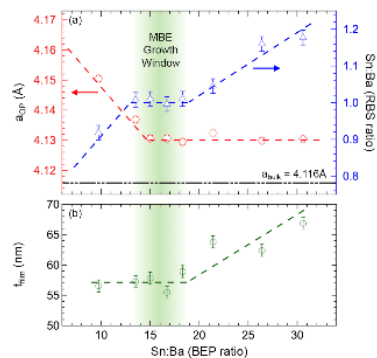


Fig. 1: Adsorption-controlled growth allows for the existence of an MBE growth window [2]

Control of the metal-insulator transition in VO₂ thin films by K deposition

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Vanadium dioxide (VO₂), which is one of the strongly correlated transition metal oxides, has widely attracted interest because of its first-order metal-insulator transition (MIT) accompanied by a structural phase transition around room temperature. Recently, it has been reported that the transition temperature (T_{MI}) of the MIT in VO₂ films is suppressed by "field-effect doping" using an electric double-layer transistor (EDLT) structure [1]. However, despite many subsequent studies, the driving mechanism of this electric-field-induced MIT still remains a controversial issue [1,2]. In order to clarify the mechanism, the direct observation of the changes in the electronic structures associated with the MIT is indispensable. However, since the sample surface is covered by an ionic liquid in the EDLT structure, it has been difficult to observe the electronic structures in the electrostatic charge-accumulated region by using photoemission spectroscopy (PES). In this study, to overcome the difficulty, we have adopted an alternative route to address the MIT induced by electrostatic electron doping [3]: we have adsorbed K atoms on the surface of VO₂ thin films, which enables us to realize the same situation of the surface carrier injection as in the case of EDLT, and observed the changes in the electronic structures across the carrier-induced MIT.

Figure 1 shows the valence band PES spectra measured at 250 K for the VO₂ thin film ($T_{\text{MI}} \sim 295$ K) before and after K deposition. Reflecting the insulating phase of VO₂ thin films, an energy gap is clearly observed at the Fermi level (E_{F}) for the bare VO₂ thin film. When K atoms are deposited on the surface of the VO₂ thin film, the clear Fermi-edge structure appears at E_{F} . These results indicate that the VO₂ thin film in insulating phase is successfully metallized by carrier injection by K deposition.

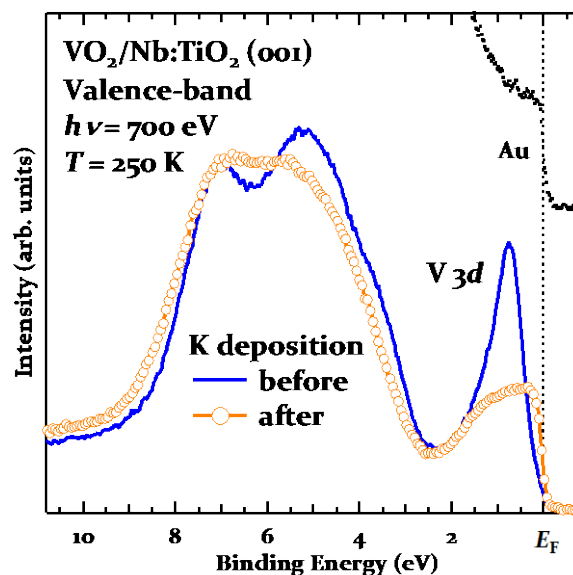


Figure 1 | Valence band PES spectra measured at 250 K for VO₂ thin films before and after K deposition.

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Role of Growth Oxygen Partial Pressure on Atomic Termination Sequence at SrRuO₃/BaTiO₃ Interface

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Since its rediscovery in the late 1980s, pulsed laser deposition (PLD) has become one of the most widely used techniques for oxide film synthesis, due to its merits of performance and simplicity. With the recent trends on miniaturizing oxide-based devices, the needs of atomic-scale control of surface/interface structure by PLD has increased. In particular, realizing uniform atomic termination sequence at surface/interface has become important for exploring novel emergent phenomena in complex oxide heterostructure.¹⁻⁴ However, selective control of surface/interface termination sequence is still challenging due to the lack of understandings on surface formation mechanism in PLD.

Here, taking the prototypical SrRuO₃/BaTiO₃/SrRuO₃ (SRO/BTO/SRO) heterostructure as a model system, we investigate the formation of different interfacial termination sequences (BaO-RuO₂ or TiO₂-SrO) with oxygen partial pressure (PO₂) during PLD. The BTO layers were grown under three background atmospheres: 1) pure oxygen atmosphere with PO₂ = 5 mTorr; 2) pure oxygen atmosphere with PO₂ = 150 mTorr; 3) argon and oxygen (O₂/Ar) mixed atmosphere with P_{O₂} = 5 mTorr and argon partial pressure P_{Ar} = 145 mTorr. Our systematic structure characterizations of these samples reveal that the total background pressure determines the plasma plume shape, growth mode, and growth rate. However, termination sequence of SRO/BTO interface is only dependent on the PO₂. These results indicate that the termination sequence at SRO/BTO interface is determined not by the plume dynamics but by the thermochemical environment conditions. Our density functional theory (DFT) calculation and phenomenological modeling also support our claim.

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Catalytically Active High-Energy Surfaces by Rapid-Anneal Solid Phase Epitaxy

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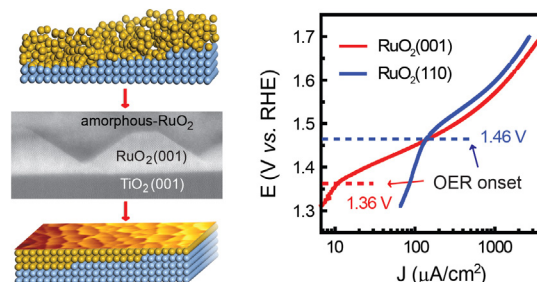
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The broad range of properties exhibited by transition metal oxides originates in their strong structure-property relations, combined with the large variety of transition metal oxides. Their functionality can be further manipulated by confining their structure into atomically precise nanoscale heterostructures. A prerequisite in this advanced tailoring is to keep the growth front flat during heterostructure growth. This has critically restricted the availability of atomically precise oxide heterostructures to crystallographic orientations with low surface energies to avoid faceting or roughening transitions.

We report a significant advance in our discovery of an extreme non-equilibrium synthesis approach for nanoscale thin films that is able to overcome this limitation. The broad applicability of this rapid-anneal solid phase epitaxial method drastically expands the family of functional nanoscale heterostructures that can be synthesized, and thus allows for a substantially broadened range of accessible properties.

We demonstrate this approach in two different functionally important transition metal oxides: TiO₂ and RuO₂. Focusing on RuO₂, a benchmark oxygen evolution catalyst, we for the first time stabilize nanometer thin, epitaxial RuO₂ films in the high surface energy (001) crystallographic orientation. We find that this novel heterostructure exhibits significantly improved oxygen evolution catalytic properties as compared to a benchmark RuO₂(110) heterostructure, quantified by their lower onset potential and higher current density, relative to that of more common RuO₂(110) films.

Non-Equilibrium Synthesis and Oxygen Evolution Reaction



Left: non-equilibrium synthesis of flat RuO₂(001) by rapid-anneal solid phase epitaxy. Right: significant reduction in the onset potential for the oxygen evolution reaction on RuO₂(001) as compared to RuO₂(110).

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Electronic Properties of Layered Iridate Epitaxial Thin-Films

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Mott insulating states featured in layered iridates, such as Sr_2IrO_4 and $\text{Sr}_3\text{Ir}_2\text{O}_7$, have attracted substantial attention due to their novel electronic states originating from strong spin-orbit coupling and electron-correlation. These iridates have been theoretically suggested as prospective compounds for new high- T_c superconducting states. However, there are still controversial issues regarding the fundamental electronic structure of these systems (e.g. strongly hybridized $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ bands [1] vs. well-separated $J_{\text{eff}} = 1/2$ and $J_{\text{eff}} = 3/2$ bands [2]).

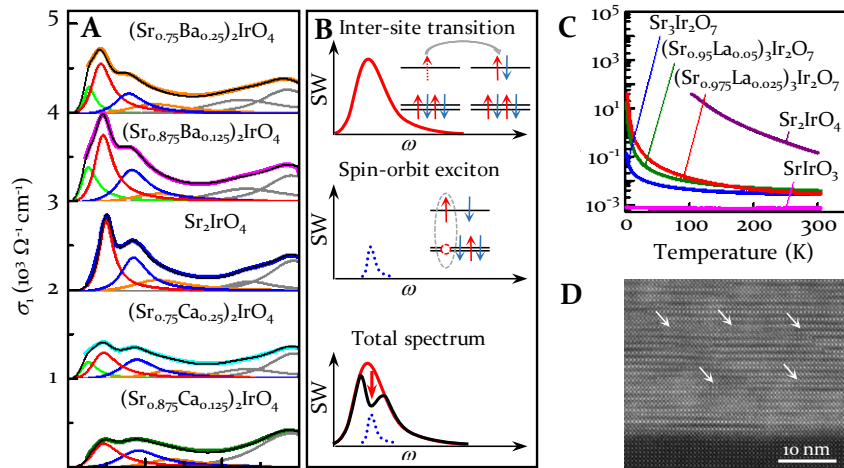
Here, we present on the electronic and optical properties of Ca- and Ba-doped Sr_2IrO_4 and La- $\text{Sr}_3\text{Ir}_2\text{O}_7$ epitaxial thin-films. The optical conductivity spectra of doped Sr_2IrO_4 shows that the increased U/W from $(\text{Sr}_{1-x}\text{Ca}_x)_2\text{IrO}_4$ to $(\text{Sr}_{1-y}\text{Ba}_y)_2\text{IrO}_4$ causes a red shift in spectral weight (A) which is due to Fano-like

coupling between inter-site $d-d$ transitions within the $J_{\text{eff}} = 1/2$ band and the spin-orbit exciton (B). Transport measurements show that the La-doped $\text{Sr}_3\text{Ir}_2\text{O}_7$ thin-films are more insulating than undoped $\text{Sr}_3\text{Ir}_2\text{O}_7$ thin-films (C). The cross-sectional scanning transmission electron microscopy (STEM) shows stacking faults, which are responsible for the increased resistivity in La-doped $\text{Sr}_3\text{Ir}_2\text{O}_7$ thin-films (D).

Our experimental results provide indispensable information in understanding the electronic properties of layered iridate thin-films.

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A) Optical spectra of doped Sr_2IrO_4 films. The experimental spectra and the fit oscillators are shown by thick and thin curves, respectively. B) Schematic of the Fano-like resonance between inter-site optical transitions and the spin-orbit exciton. C) T -dependent resistivity of the $(\text{Sr}_{1-x}\text{La}_x)_3\text{Ir}_2\text{O}_7$ films. D) STEM image of $\text{Sr}_3\text{Ir}_2\text{O}_7$ film. Arrows show some areas with stacking fault.

TwinBeam PLD - new possibilities for advanced functional films

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At present the growth of thin films by using the PLD technique is limited to the availability of dense ceramic targets of the stoichiometric compounds. Some phases do not withstand the sintering conditions and therefore poorly sintered or composite targets are used instead. This may bring some problems of particle ejection and composition deviations in the growing film. Another typical problem in PLD arises when some element in the compound has a higher volatility (Pb, Bi, etc), which induces serious deviations in the film composition. This is normally compensated by preparing targets enriched in the volatile element, which needs to be adjusted for each deposition conditions. These limitations can be overcome by the use of simultaneous ablation of two targets to composite the final film material.

The new SURFACE Twin Beam PLD technology makes use of a dual laser beam of the same laser, separated by a laser beam splitter and a new target manipulator that combines three standard single targets (2" diameter) ablation with a double target (2x1" diameter) position for the simultaneous ablation. The angle of the twin targets can be adjusted from a confocal to a cross beam configuration.

This new technology does not need a new PLD system, it is an available upgrade for all SURFACE PLD-Workstation systems. The new TwinBeam option includes only an upgrade of the standard target manipulator to the TwinBeam version. The necessary additional beam line is an extension to the existing one and the tuning of the fluence will be done with two manual or motorized laser beam attenuators. A software upgrade incorporates this new feature in the highly flexible PLUMEmaster user interface.

New process features are integrated and fully software controlled:

- software controlled in-situ fluence variation between the two targets
- fluence modulation of one beam synchron to the RHEED oscillation of the film growth
- synchronic variation of the plume angle at TwinBeam targets from a cross-beam¹⁾ to confocal plume to parallel plume orientation
- enhanced combinatorial features:
 - + in-situ composition spread deposition (ICSD) of two materials
 - + better material mix with no limitation of film thickness and no post-annealing step
 - + large fluence differences and modulation possible to combine metals with oxides

The TwinBeam option extends the flexibility of the SURFACE PLD workstation and is a powerful support of the SURFACE wedge heater which allows other combinatorial operations like wedge shaped films or deposition through shadow masks.

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Reversible spin texture in ferroelectric HfO₂

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Spin-orbit coupling effects occurring in non-centrosymmetric materials are known to be responsible for non-trivial spin configurations and a number of emergent physical phenomena. Ferroelectric materials may be especially interesting in this regard due to reversible spontaneous polarization making possible for a non-volatile electrical control of the spin degrees of freedom. Here, we explore a technologically relevant oxide material, HfO₂, which has been shown to exhibit robust ferroelectricity in a non-centrosymmetric orthorhombic phase. Using theoretical modelling based on density-functional theory, we investigate the spin-dependent electronic structure of the ferroelectric HfO₂ and demonstrate the appearance of chiral spin textures driven by spin-orbit coupling. We analyze these spin configurations in terms of the Rashba and Dresselhaus effects within the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian model and find that the Rashba-type spin texture dominates around the valence band maximum, while the Dresselhaus-type spin texture prevails around the conduction band minimum. The latter is characterized by a very large Dresselhaus constant $\alpha_D = 0.578 \text{ eV \AA}$, which allows using this material as a tunnel barrier to produce tunneling anomalous and spin Hall effects that are reversible by ferroelectric polarization.

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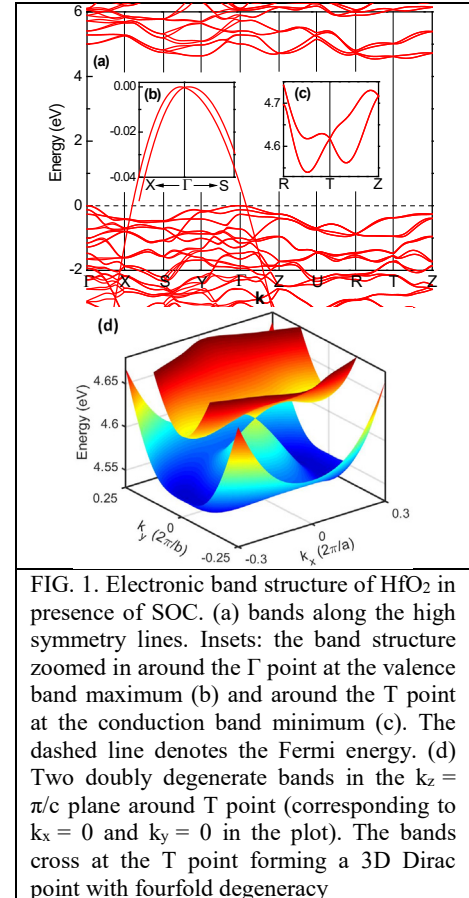


FIG. 1. Electronic band structure of HfO₂ in presence of SOC. (a) bands along the high symmetry lines. Insets: the band structure zoomed in around the Γ point at the valence band maximum (b) and around the T point at the conduction band minimum (c). The dashed line denotes the Fermi energy. (d) Two doubly degenerate bands in the $k_z = \pi/c$ plane around T point (corresponding to $k_x = 0$ and $k_y = 0$ in the plot). The bands cross at the T point forming a 3D Dirac point with fourfold degeneracy

Internal Charge Transfer and Hopping Conduction in MBE-Grown $\text{SrTiO}_3/\text{Nd}_x\text{Ti}_{1-x}\text{O}_3/\text{SrTiO}_3$ Heterostructures

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Interface between a polar and a nonpolar insulating perovskite oxide such as $\text{LaAlO}_3/\text{SrTiO}_3$ has created a lot of excitement due to the discovery of interface conductivity, magnetism, 2D superconductivity and Rashba-like spin-orbit interactions. Consensus however has not yet been reached concerning the mechanism(s) of interfacial conduction.

In this talk, we will present detailed study from experiments and modeling that carrier densities much higher than expected based on resolution of the polar discontinuity at perovskite oxide heterojunctions can be achieved via band engineering and internal charge transfer. The SrTiO_3 (8 u.c.) / NdTiO_3 (2 u.c.) / SrTiO_3 (8 u.c.) / LSAT(001) heterostructure shows the expected electronic reconstruction behavior starting at $t = 2$ u.c., but then exhibits a higher carrier density regime at $t \geq 6$ u.c. due to additional charge transfer from band alignment [1].

Furthermore, consistent with the electronic reconstruction model, we show that the carrier density of the two-dimensional electron gas (2DEG) in SrTiO_3 (8 u.c.) / $\text{Nd}_{1-x}\text{TiO}_3$ (2 u.c.) / SrTiO_3 (8 u.c.) / LSAT(001) can be modulated over a wide range through systematic control of the Nd vacancies [2]. Samples with large amount of Nd-vacancies turned insulating with the electronic transport being governed by the Mott VRH near the metal-insulator boundary, crossing over to an ES VRH mechanism with increasing Nd-vacancies and disorder (see Fig. 1). Combining DFT modeling and experiments using x-ray photoelectron spectroscopy, scanning transmission electron microscopy, electron energy loss spectroscopy, energy dispersive x-ray spectroscopy and electronic transport measurements, we will discuss the origin of these carriers, dimensionality and different transport mechanisms.

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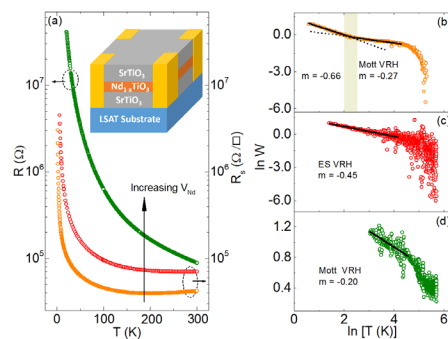


Fig 1(a). Temperature dependence of the resistance (left axis) and sheet resistance (right axis) of the three samples with different Nd-vacancies. (b-d) The corresponding temperature dependence of the reduced activation energy using the Zhabrinskii analysis.

Electronic Reconstruction Enhanced Tunneling Conductance at Terrace edges of Ultrathin Oxide Films

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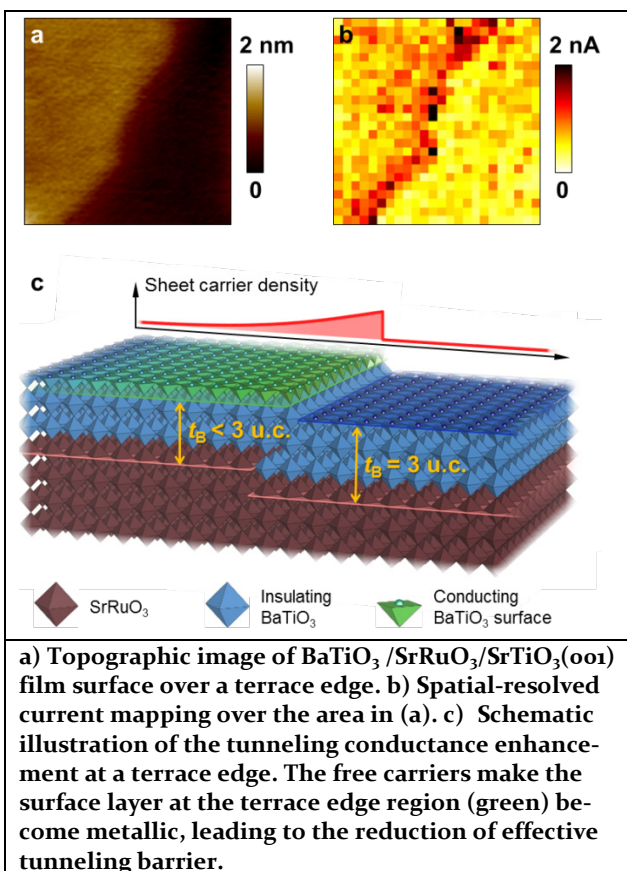
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One of the fundamental fruitings in quantum mechanics is that electrons can tunnel across an insulating barrier when it is few-nanometers-thick. During the last two decades, due to the rapid advancements in heteroepitaxy growth techniques, reducing the oxide film thickness to nanometer scale while retaining its original ferroic order becomes feasible. Such capabilities enable oxide-based novel tunneling devices with superior data storage capability, including magnetic spin filters and ferroelectric/multiferroic tunnel junctions. In the few-nanometer-thick epitaxial oxide systems, atomic-scale structural imperfections, such as the ubiquitously existed one-unit-cell-high terrace edges, can dramatically affect the tunneling probability and device performance. However, the underlying physics has not been investigated adequately.

Here, taking ultrathin BaTiO₃ films as a model system, we report an intrinsic tunneling conductance enhancement near the terrace edges. Spatial-resolved current-voltage spectroscopy demonstrates the existence of highly-conductive regions (tens of nanometers-wide) near the terrace edges. First-principles calculations suggest that the terrace edge geometry can trigger an electronic reconstruction, which reduces the effective tunneling barrier width locally. Furthermore, such tunneling conductance enhancement can be discovered in other transition-metal-oxides and controlled by surface termination engineering. The controllable electronic reconstruction could facilitate the implementation of oxide electronic devices and discovery of exotic low-dimensional quantum phases.



Growth of High Quality Epitaxial LaAlO₃ on SrTiO₃ on (001) Si via Molecular-Beam Epitaxy

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We report the *in-situ* integration of epitaxial LaAlO₃ on silicon using a 5 unit-cell (u.c.) thick SrTiO₃ buffer layer. Both LaAlO₃ and SrTiO₃ films were grown with molecular-beam epitaxy (MBE). Strontium and titanium molecular beams were generated using a conventional effusion cell and a Ti-Ball™ titanium sublimation pump, respectively. Lanthanum and aluminum beams were generated via conventional effusion cells. The 5 u.c. thick SrTiO₃ buffer layer was grown using the so-called epitaxy-by-periodical-annealing method. To ensure the stoichiometry of the LaAlO₃ film, lanthanum and aluminum fluxes were calibrated by codeposition of LaAlO₃ for stoichiometric reflection high-energy electron diffraction (RHEED) patterns and also by observing the RHEED oscillations for the growth rate. After the calibration, LaAlO₃ was grown by codeposition with an oxygen partial pressure of $\sim 2 \times 10^{-7}$ Torr at the substrate temperature of ~ 590 °C.

In-situ RHEED images showed both the SrTiO₃ and LaAlO₃ films were smooth. *Ex-situ* X-ray diffraction (XRD) θ -2 θ scan shows only *o*o*l* reflections of LaAlO₃, indicating the film is epitaxial. A ϕ scan shows the LaAlO₃ layer on silicon to have a cube-on-cube epitaxial orientation relationship. The rocking curve of the LaAlO₃ layer shows a single peak with a full width at half maximum (FWHM) of 0.02°, which is comparable to single crystal LaAlO₃ substrates and is the narrowest LaAlO₃ rocking curve ever reported for the growth of LaAlO₃ on silicon.

Using MBE, our *in-situ* growth yields a high quality epitaxial LaAlO₃ film on (001) Si, which could find its applications in serving as a twin-free template for multi-functional oxide integration on silicon.

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Carrier Density Modulation over an Exceptional Voltage Window in BaSnO₃ Films via Ion Gel Gating

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BaSnO₃ has drawn interest recently due to its outstanding room temperature mobility and potential applications in power electronics, oxide transistors, transparent conductors, etc. Here we report effective control of the electronic transport properties of sputtered oxygen-vacancy doped BaSnO₃ (BaSnO_{3-δ}) films via ion gel gating in electric double layer transistor structures. The electron density of the starting films is tuned by thickness, from $4 \times 10^{19} \text{ cm}^{-3}$ at 12 nm to much lower densities at lower thickness. The response to gate voltage is found to be notably robust, with largely reversible response (even in vacuum) over an exceptionally wide window from -4 to +4 V, even at 300 K. The data support predominantly electrostatic response, unlike many other oxides, which we ascribe to Sn redox stability. In this manner the sheet resistance of 12-nm-thick BaSnO₃ films can be modulated over a factor of 200 at 300 K. Hall measurements show that the maximum induced carrier density is $8 \times 10^{13} \text{ cm}^{-2}$ at 4 V and that mobility increases monotonically with carrier density from below $1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ to $18 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ due to charged defects as dominant scattering centers. Similar measurements at lower thickness/carrier density will also be discussed and show larger modulation of resistance. Finally, Hall measurements along with a two-conduction-channel analysis reveal a 50% increase in mobility in an ion-gel-gated La-doped BaSnO₃ film grown by molecular beam epitaxy, with a starting room temperature mobility of $66 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, suggesting ion gel gating as an effective method of improving BaSnO₃ mobility.

The Effect of Fluorination Conditions on the Physical Properties of $\text{SrMnO}_{3-\delta}\text{F}_\gamma$ Epitaxial Films

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Controlling the carrier concentrations in electronic materials through doping is of fundamental importance in engineering physical properties for applications in electronic devices. In ABO_3 perovskites, altering the electron count on the B -site cation is typically achieved through A -site cation substitution. Less explored is anion substitution; for example, F^- substitution for O^{2-} provides an alternative means to tailor the nominal B -site valence, the iconicity of the B -O bond, and the lattice volume. While bulk oxyfluorides have received interest over the last two decades, epitaxial perovskite oxyfluorides have only recently been reported.^[1,2] In this work, we describe topotactic fluorination reactions in $\text{SrMnO}_{3-\delta}\text{F}_\gamma$ epitaxial perovskite films and the resultant effects on physical properties. The fluorination process is carried out through thermal decomposition of fluorine-containing polymers as a fluorine source to react with as-grown oxide films. Three different polymers with different F:H ratio were utilized: PVDF ($\text{C}_2\text{H}_2\text{F}_2$), PTFE (C_2F_4), and PVF ($\text{C}_2\text{H}_3\text{F}$), allowing for the impact of the fluorine source material on the incorporated F content (γ) to be determined. The F concentration and its depth-dependent uniformity and the crystalline quality of the $\text{SrMnO}_{3-\delta}\text{F}_\gamma$ films, as probed by X-ray photoemission spectroscopy and X-ray diffraction, are found to depend strongly on the fluoropolymer and the reaction temperature. The electronic, magnetic and optical behavior of $\text{SrMnO}_{3-\delta}\text{F}_\gamma$ fluorinated films, SrMnO_3 oxidized films and $\text{SrMnO}_{2.5}$ as-grown films will be compared to illustrate the potential of anion doping to engineer properties.

The work is supported by the National Science Foundation (CMMI-1562223).

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Visualizing ballistic phonon transport in ferroelectric BaTiO₃ upon localized terahertz field excitation

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Controlling complex oxides on ultrafast time scales using electric fields is an essential step towards realizing ultrafast oxide electronics. The recent development of intense terahertz (THz) sources allows researchers to apply ultrafast ultrahigh electric bias without forming contacts with metal electrodes. The THz field can directly couple into electric polarization of ferroelectric oxides, providing the basis for ultrafast control. Here we report the use of single-cycle, sub-ps duration THz pulses further enhanced by a metasurface antenna, with a localized peak electric field on the order of MV/cm, to control the structural properties of a ferroelectric BaTiO₃ film. Using time-resolved x-ray diffraction microscopy, we image the evolution of localized strain profile with sub- μm and sub-ns spatiotemporal resolution. We show that the selective THz excitation of low-frequency phonons significantly enhances the range of in-plane energy dissipation via a mechanism associated with ballistic phonon transport over a distance of hundreds of nm, two orders of magnitude larger than the frequency-averaged phonon mean free path. On longer length scales, the phonon transport becomes diffusive and heat transport into the substrate dominates the recovery of the THz-induced transient strain. The ballistic to diffusive heat transport dynamics across the nano-to-mesoscale are reproduced by the molecular dynamic calculations and diffusive modeling respectively. This real-time real-space visualization of THz-matter interaction opens opportunities to image engineered nanoscale transient structural states for new functionalities.

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Transport in Dual-gated Nb:SrTiO₃ Quantum Wells

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Dual-gating is a powerful technique to tune the carrier density and mobility of two-dimensional electron systems. In particular, tunability is effective in SrTiO₃ thin films because the SrTiO₃ has a large lattice dielectric constant, reaching to 20,000 at cryogenic temperature. However, this phenomena on the other hand are known to cause an enormous surface depletion in a thin n-type SrTiO₃ film, giving rise to a reduced carrier density concomitant with low mobility.[1] Remarkably, sandwiching the thin doped layer with undoped SrTiO₃ cap and buffer layers, in a delta-doped structure, allows us to avoid these drawbacks, resulting in high mobility channels.[2-4] This delta-doped SrTiO₃ heterostructure is known to have several distinctions from the LaAlO₃/SrTiO₃ interface: it is a symmetric quantum well, in the absence of strong interfacial disorder or local ferromagnetism.[5] Moreover, the delta-doped structures allow systematic variation in their transport properties by controlling the structural parameters such as cap layer thickness, doped layer thickness, and doping concentration.

Here, we present how the transport properties of delta-doped Nb:SrTiO₃ structure are controlled by electrostatic dual-gating. Fitting the magneto-resistance curve allows us to extract the inelastic scattering time and the spin-orbit scattering time. We show that tuning these characteristic scattering times result in a weak localization to weak anti-localization crossover.

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Tuning Spin Relaxations in Ultrathin Epitaxial SrIrO_3 Thin Films via Ferroelectric Gating

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As the end member ($n = \infty$) of the Ruddlesden-Popper series ($\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$), SrIrO_3 (SIO) remains paramagnetic, semimetallic and with nontrivial topological properties. Its strongly correlated nature and large spin orbital coupling (SOC) make SIO a promising material candidate for spintronic applications. In this work, we have carried out systematic studies on the transport properties of epitaxial SIO thin films with thickness of 1.6 – 21.2 nm grown on SrTiO_3 (STO) substrates. While the thick films are metallic with moderate resistance change, the films in the 2D regime exhibit an upturn at low temperature, which is attributed to weak localization (WL) and electron-electron interaction. As a feature of strong SOC, weak anti-localization (WAL), manifested as negative quantum interference correction to the conductance, has been observed in the magnetoresistance (MR) at low temperatures (2 – 5 K). Using the Maekawa-Fukuyama model, we have extracted the inelastic scattering length and spin precession length, which show distinct temperature dependences. Moreover, the linear mobility dependence of spin precession time points to Elliott-Yafet mechanism as the dominant spin relaxation mechanism.

By fabricating the $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT)/SIO (1.6 – 2.8 nm) heterostructures, we have demonstrated nonvolatile resistance change in SIO via switching the polarization field of PZT. We extracted the carrier density and mobility based on the Hall effect measurements combined with the MR, which reveals tuning of the nearly compensated electron- and hole- densities. The onset temperature of the resistance upturn is also modulated by the ferroelectric field effect, with an up to 10 K shift observed. The modulation of MR indicates the change of SOC strength in SIO. Our results demonstrate a feasible way to manipulating the SOC at the interface of all oxides heterostructures.

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Lithium Intercalation into $\text{La}_{2/3}\text{TiO}_3$ and $\text{La}_{1/3}\text{NbO}_3$ thin films

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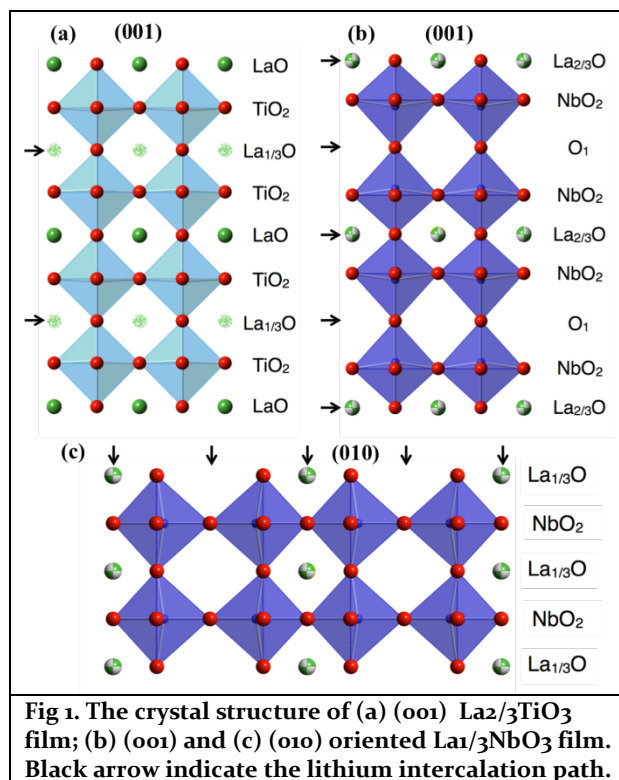
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Lithium-lanthanum-titanate ($\text{La}_{(2/3)-x}\text{Li}_x\text{TiO}_3$, LLTO) and Lithium-lanthanum-niobate ($\text{La}_{(1/3)-x}\text{Li}_x\text{NbO}_3$, LLNb) are promising perovskite-type electrolytes for solid-state batteries and other oxide-based electrochemical devices. The high lithium ion conductivity of these perovskite electrolytes is considered to be due to large A-site vacancies concentration, allowing the motion of lithium ions by a vacancy mechanism.¹ However, one major challenge for these perovskite electrolytes is the instability against Li anode.¹ Motivated by this, our experiments are designed to study how lithium intercalate into LLTO and LLNb and the influence from A-site vacancies and film orientation.

Hetero-epitaxial $\text{La}_{2/3}\text{TiO}_3$ and $\text{La}_{1/3}\text{NbO}_3$ thin films (fully delithiated version of LLTO and LLNb) are deposited on SrTiO_3 single-crystal substrate using layer-by-layer pulsed laser deposition (PLD). In this process, alternating layers of La_xO and $\text{TiO}_2/\text{NbO}_2$ are deposited and the A-site vacancies can be easily tuned by controlling the amount of La in La_xO layer. After deposition, a bias voltage is applied (using a lithium anode) to intercalate lithium into the film, during which in-situ XPS and electrical measurements are carried out to monitor the change in valence state and the electronic conductivity.

By changing the La concentration in La_xO layer during film growth, the orientation of the film can be changed from (001) to (010) as indicated in Fig.1 (b) and (c). It is expected that these two films will have very different lithium intercalation rates. There are fewer lithium intercalation paths through (001) film as the A-site deficient planes are parallel to film surface. In contrast, the lithium ion can easily penetrate into the (010) film because all paths are normal to surface.



This study will provide in-depth understanding of interaction between lithium and A-site cation deficient perovskites. Furthermore, defect engineering of the cation sublattice may prove to be an effective means of tuning the properties of future oxide electronic devices.

References:

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