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Oxide 2DEG for spin-charge interconversion

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Spintronics traditionally relied on ferromagnetic metals as spin generators and detectors. A new approach, spin-orbitronics, exploits the interplay between charge and spin currents enabled by the spin-orbit coupling in non-magnetic systems.

We studied the potential of bidimensional electron gases (2DEG) that form at the interface between LaAlO₃ and SrTiO₃ [1] or by deposition of Al, Ta or Y on SrTiO₃ [2], Ca:SrTiO₃ [3] or KTaO₃ [4] for spin-current to charge-current conversion [2, 3, 4, 5]. The sizeable Rashba spin-orbit coupling of the gas [6] allows to obtain very efficient conversions, larger than those of the topological insulator α -Sn or the Bi/Ag interface [7]. This conversion efficiency can be highly modulated by a gate voltage and linked to the band structure [2]. We also studied the charge to spin conversion, detected through simple magnetotransport experiments [8, 4].

Moreover, we exploited the electric-field induced ferroelectricity in SrTiO₃ to manipulate the spin-orbit properties of a 2DEG and convert spin currents into either positive or negative charge currents in a non-volatile manner [9].

This suggests that oxide interfaces [10] have a strong potential for spin-based information readout in novel memory [11] or for a new generation of spin-based devices, in which non-volatility would be provided by ferroelectricity rather than by ferromagnetism.

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Interface Charge Engineering in Ferroelectric-Gated Mott Transistors

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Epitaxial complex oxide heterostructures composed of ferroelectric gates and correlated channels (Fig. 1a) are promising material platforms for developing high performance, energy efficient electronic devices. In this study, working with a ferroelectric $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$ (PZT) gate, we present a systematic study of the ferroelectric field effect in rare earth nickelates $\text{Sm}_{0.5}\text{Nd}_{0.5}\text{NiO}_3$ (SNNO), NdNiO_3 (NNO), and LaNiO_3 (LNO) to identify the optimal channel materials for constructing nonvolatile Mott transistors. For single layer nickelate channels, the resistance switching ratio $\Delta R/R_{\text{on}}$ peaks near the electrical dead layer thickness, then decreases abruptly due to strong depolarization. The highest room temperature $\Delta R/R_{\text{on}}$ of $\sim 200\%$ is observed in 1 nm NNO and LNO channels due to their small electrical dead layer thickness. We also show that inserting a $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) buffer layer can lead to over two orders of magnitude increase in $\Delta R/R_{\text{on}}$ for devices with the same channel thickness (Figs. 1b-c), which is attributed to the tailored carrier density profile due to the interfacial charge-transfer between nickelate and LSMO. A record high $\Delta R/R_{\text{on}}$ of 9,000% is observed in the 1.2 nm LNO/0.4 nm LSMO bilayer channel at 300 K (Fig. 1c). Our study addresses one of the key material challenges that limit the application potential of epitaxial ferroelectric-gated Mott transistors.

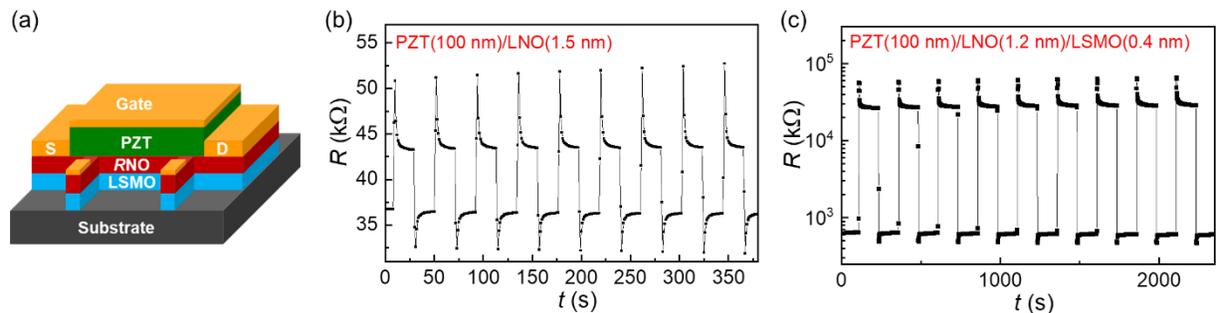


Figure 1 (a) Device schematic. (b) Resistance switching in a 1.5 nm LNO channel at 300 K. (c) Resistance switching in a 1.2 nm LNO/0.4 nm LSMO channel at 300 K.

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Large topological Hall effect and spin textures in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ / SrIrO_3 bilayers

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Topologically protected spin textures, such as magnetic skyrmions, have attracted considerable interest in recent years, as they can be used as information vectors in low-power information technologies. Spin textures can be stabilized by the interplay between exchange interaction, promoting parallel spin alignment, and Dzyaloshinskii-Moriya interaction, an antisymmetric exchange interaction resulting from the combination of inversion symmetry breaking and strong spin-orbit coupling, which twists spin directions. Although skyrmion or skyrmion bubbles have been observed in non-centrosymmetric single magnetic oxide layers [1], interfaces naturally supply the symmetry breaking and favor the observation of spin textures in heterostructures [2,3]. The non-trivial topological charge of the skyrmions gives rise to an antisymmetric contribution to the transverse resistivity, the so-called topological Hall effect, THE. Whether THE results from spin chirality or from magnetic inhomogeneity has become an actively debated matter [4,5], boosted by the fact, that, frequently, reports of the THE are not accompanied by real space imaging of the magnetic textures. In this work we report large THE in bilayers combining ultrathin $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ and strong spin orbit SrIrO_3 in a wide temperature range. Real space observations of the magnetic textures by magnetic force microscopy (MFM) and photoemission microscopy using x-ray magnetic circular dichroism contrast (PEEM) evidence a coarsened small grain (100 nm) magnetic structure inferred from image analysis, consistent with the nucleation and clustering of skyrmions or skyrmion bubbles. Field cool in strong magnetic fields above 5T suppress the THE signal and consistently the MFM contrast of the magnetic textures, suggesting the possibility to control the skyrmions. Magnetometry and anomalous Hall effect will be discussed to infer the relative importance of magnetic inhomogeneity.

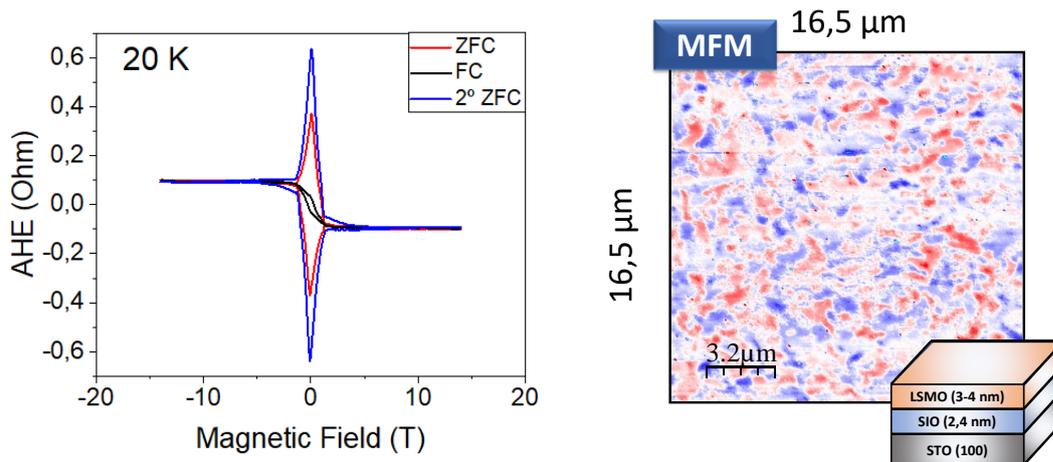


Figure 1. Left panel. Anomalous Hall effect measured at 20 K after field cooling (FC) (black line) and zero field cooling (ZFC) (red and blue lines). Right panel. MFM image at 17 K after ZFC.

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Charge-transfer engineering at polar double-perovskite/perovskite interfaces

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Interfacial charge-transfer phenomena are a successful pathway to engineering novel functionalities that are absent in bulk. However, they can also deteriorate the properties of interest especially, in the ultrathin limit. Advanced approaches to manipulate the electronic state of a buried interface are thus very intriguing.

Here we demonstrate that at the interface between the double perovskite $\text{La}_2\text{NiMnO}_6$ (LNMO) and SrTiO_3 (STO) (Fig. 1a), a charge transfer triggered by a polarity mismatch emerges. LNMO is a ferromagnetic insulator with a bulk Curie temperature (T_C) around 280 K, resulting from positive superexchange between the long-range-ordered Ni^{2+} and Mn^{4+} cations. Bulk-like behavior is reproduced in high-quality epitaxial films [1], and films as thin as at least 2-3 unit cells still show ferromagnetic behaviour. However, the polarity-induced electronic reconstructions (see Mn^{3+} emergence in Fig. 1d) result in a reduction of T_C and magnetization of the ultrathin films further than merely dimensionality effects (Fig. 1e).

To restore the correct electronic configuration of the buried LNMO//STO interface, we design a top-interface engineering approach through a LaNiO_3 (LNO) overlayer. The presence of LNO is fundamental in displacing the interfacial charge and allows to reestablish the ferromagnetism in LNMO. The LNO-induced electron migration (supported by XAS, XMCD, TEM and DFT+U) is very general, being also obtained on polar LNMO//LSAT interfaces, and can be potentially exploited to tune the properties of other polar oxide interfaces.

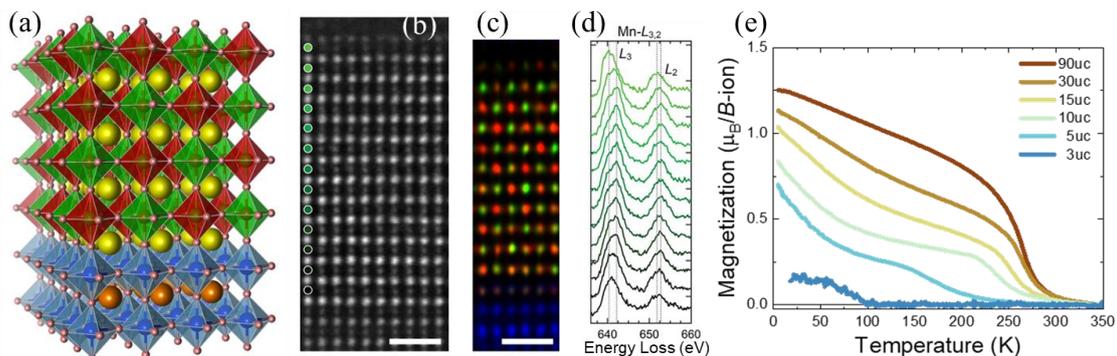


Figure 1. (a) Sketch of the double perovskite/perovskite LNMO//STO heterostructure. (b) HAADF image of a 13 uc LNMO film. (c) STEM-EDX map showing the distribution of Mn (green), Ni (red) and Ti (blue). Same colours are used in a). (d) EELS of Mn $L_{3,2}$ edges. (e) Magnetization vs temperature for various LNMO film thicknesses.

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Optimizing Monolayer Ferroelectric ZrO₂ on Si (001)

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A single atomic layer of ZrO₂ on Si (001) shows ferroelectric switching behavior. [1] Being at the ultimate thickness limit of a monolayer (ML), this system has the potential to aggressively scale down device size. Motivated by density functional theory (DFT) calculations that attribute the polarization switching to displacements of Zr-O dipoles, we aim to control the magnitude of capacitance – voltage (CV) hysteresis by controlling the density of Zr-O dipoles in the ML ZrO₂ film. The Zr-O dipole density is controlled through film annealing conditions characterized using x-ray photoelectron spectroscopy (XPS) measurements of Zr 3d core level energies, which are sensitive to Zr valence. Additional synchrotron x-ray absorption spectroscopic measurements also indicate changes in Zr valence as the process conditions are changed. The CV measurements show that the magnitude of the hysteresis is proportional to the density of Zr-O dipoles controlled by thin film annealing conditions, with Zr⁴⁺ XPS relative intensity ranging from about 30% to 90%. The magnitude of hysteresis in the CV changes from about 0.1 to 0.5 V and correlates directly with Zr⁴⁺ content.

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Strange metal states and quantum criticality in doped $J_{eff}=1/2$ Mott states

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$J_{eff}=1/2$ Mott insulators are born from the delicate interplay between strong spin-orbit coupling, crystal field splitting, and on-site Coulomb interactions most often found in $4d$ and $5d$ transition metal oxides. Both the single layer ($n=1$) and bilayer ($n=2$) members of the strontium iridate Ruddlesden-Popper family of $Sr_{n+1}Ir_nO_{3n+1}$ compounds form this Mott state with spin-orbit entangled wave functions and are predicted to support correlated metallic states and various forms of emergent order when doped away from half-filling. Here I will present some of our recent progress in understanding how these parent $J_{eff}=1/2$ Mott states transition into nearby correlated metal states and whether a quantum critical phase transition results. Particular focus will be given to understanding the effect of Ru-alloying and “hole-doping” on the metal-to-insulator and magnetic phase transitions in single layer and bilayer compounds as they are driven from their iridate (Sr_2IrO_4 , $Sr_3Ir_2O_7$) to ruthenate endpoints (Sr_2RuO_4 , $Sr_3Ru_2O_7$).

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Machine-Learning Assisted Quantum Materials Discovery And Optimization:

Metal-Insulator Transition Compounds

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Metal-insulator transition (MIT) compounds are materials that may exhibit metallic or insulating behavior, depending on the physical conditions, and are of interest due to their correlated properties as well as due to their potential applications in emerging microelectronics. Nonetheless, only around 60 materials with a thermally-driven metal-insulator transition are known, and their computational discovery is difficult due to the non-equilibrium nature of the transition, and the complexity of the many-body problem. To address this issue, we have built the first database of all known thermally-driven metal-insulator transition compounds, as well as stoichiometrically related compounds, and a machine-learning based classifier tool to accelerate their discovery - and provided both to the wider public, with no installation required. Using Bayesian optimization techniques, we have also optimized specific materials families to find materials which are both stable, and may display strong MIT behavior while minimizing the number of calculations. Time permitting, we will present possible new metal-insulator transition oxide compounds identified through a combination of machine learning and DFT calculations, as well as preliminary experimental results on their experimental validation.

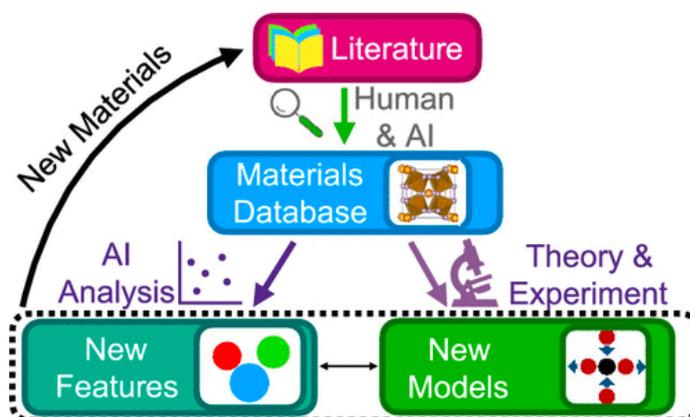


Figure 1. Incorporation of Machine Learning into the process of discovery and understanding of novel quantum materials.

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Growth of PdCoO₂ films with controlled termination by MBE and determination of their electronic structure by ARPES

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Utilizing the powerful combination of molecular-beam epitaxy (MBE) and angle-resolved photoemission spectroscopy (ARPES) we produce and study the effect of different terminating layers on the electronic structure of the metallic delafossite PdCoO₂. Attempts to introduce unpaired electrons and synthesize new antiferromagnetic metals akin to the isostructural compound PdCrO₂ have been made by replacing cobalt with iron in PdCoO₂ films grown by MBE. Using ARPES, we observe similar bulk bands in these PdCoO₂ films with Pd-, CoO₂-, and FeO₂-termination. Nevertheless, Pd- and CoO₂-terminated films show a reduced intensity of surface states. Additionally, we are able to epitaxially stabilize PdFe_xCo_{1-x}O₂ films which show an anomaly in the derivative of the electrical resistance with respect to temperature at 20 K, but do not display pronounced magnetic order.

Metal insulator transition in ^{18}O isotope substituted Vanadium dioxide

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Vanadium dioxide VO_2 is a transition metal oxide showing a metal-to-insulator transition (MIT) close to room temperature, at around 340 K. The decrease in electrical conductivity is accompanied by a structural change from monoclinic to a rutile lattice. Despite its discovery in the 1960s the origin of the phase transition is still debated and scenarios based on electron-lattice correlations (Peierls transition) or electron-electron correlations (Mott transition) have been proposed. In order to explore the effect played by phonons in this phase transition, we studied the MIT in isotope-substituted V^{18}O_2 .

We synthesized V^{16}O_2 and V^{18}O_2 microcrystals. XRD analysis shows that the crystal structure is identical for both isotopes. Raman spectroscopy and transport measurements show a clear lowering of phonon energies (1-3%) and a 1.5 K MIT temperature increase (0.5%) for the V^{18}O_2 samples.

This shift can be accounted for by calculating the phonon free energy reduction, which is larger for the monoclinic insulating phase. Our results underline the importance of lattice dynamics in setting the MIT transition temperature and indicate that electronic and lattice degrees of freedom are strongly intertwined.

Magnon Hanle experiments in antiferromagnetic α -Fe₂O₃

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Antiferromagnetic materials host pairs of spin-up and spin-down magnons as the quantized spin excitations. We describe them in terms of a magnonic pseudospin [1]. The close analogy to the spin of charge carriers leads us to the prediction of novel fascinating magnon transport phenomena [1,2,3]. We observe and investigate these effects in the electrically insulating oxide α -Fe₂O₃ (hematite). In its antiferromagnetic phase below 953 K, α -Fe₂O₃ shows a finite Dzyaloshinskii-Moriya interaction, giving rise to a slight canting of the antiferromagnetic sublattice magnetizations in the magnetic easy (0001) plane at room temperature and, hence, to a residual net magnetic moment [4,5]. The combination of α -Fe₂O₃ thin films and heavy metal Pt electrodes exhibiting a large spin-orbit coupling represents a prototypical bilayer system to investigate electrical magnon injection and detection [2,3], spin-Hall magnetoresistance [6], as well as antiferromagnetic magnon propagation [1,2,3].

We study the diffusive spin transport in a non-local geometry in two-terminal devices, each consisting of two Pt electrodes on α -Fe₂O₃ thin films with different thicknesses. Using a current reversal method enables us to discriminate between effects of electrical and thermal origin. We demonstrate the electrical magnon injection, diffusive magnon transport, and magnon detection [2,3]. We observe the coherent precession of the magnonic pseudospin caused by the easy-plane anisotropy and the Dzyaloshinskii-Moriya interaction [2,3]. Applying an external magnetic field allows us to control the precession frequency and, hereby, to interpret our observation as the magnonic analogue of the electronic Hanle effect [2,3]. In "thick" films, we additionally observe an oscillating behavior of the magnon spin signal in the high magnetic field range as well as an offset signal in the low magnetic field regime [3]. We attribute this offset signal to the presence of finite-spin low-energy magnons [3]. Our findings unlock the high potential of antiferromagnetic magnonics towards the realization of electronics-inspired phenomena.

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Superconductivity in a quintuple-layer square-planar nickelate

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Since the discovery of high-temperature superconductivity in copper oxide materials, there have been sustained efforts to both understand the origins of this phase and discover new cuprate-like superconducting materials. One prime materials platform has been the rare-earth nickelates; indeed, superconductivity was recently discovered in the doped compound $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ [1]. Undoped NdNiO_2 belongs to a series of layered square-planar nickelates with chemical formula $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$ and is known as the ‘infinite-layer’ ($n = \infty$) nickelate. Here, we report the synthesis of the quintuple-layer ($n = 5$) member of this series, $\text{Nd}_6\text{Ni}_5\text{O}_{12}$, in which optimal cuprate-like electron filling ($d^{8.8}$) is achieved without chemical doping [2]. We observe a superconducting transition beginning at ~ 13 K. Electronic structure calculations, in tandem with magnetoresistive and spectroscopic measurements, suggest that $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ interpolates between cuprate-like and infinite-layer nickelate-like behaviour. By engineering a distinct superconducting nickelate, we identify the square-planar nickelates as a new family of superconductors that can be tuned via both doping and dimensionality.

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Pauli-limit violation in nickelate: to what extent a high- T_c cuprate analogue?

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The discovery of high-temperature (high- T_c) superconductivity in cuprate four decades ago motivates intense theoretical and experimental efforts to pursue and understand the phenomenon [1]. One of the ideal routes is through a ‘twin sister’ – a cuprate analogue which mimics the electronic and structural templates of the high- T_c cuprate. Standing beside copper in the periodic table, Ni¹⁺ in infinite-layer phase hosts $3d^9$ electronic structure with lifted orbital degeneracy resembles Cu²⁺ state in cuprate superconductors. Despite more than two decades of theoretical predictions [2,3], superconducting infinite-layer nickelate was only successfully synthesized in 2019 in thin film form [4]. Since then, considerable advancements in both theoretical and experimental studies of this newfound long-promise nickelate superconductor have been made. Following the two decades theoretical debates, the recent experimental data suggests both significant similarities and distinctions to the high- T_c cuprate [5-9]. We will discuss these aspects along with our recent magnetotransport data measured in magnetic fields up to 55 T and at temperature down to 30 mK that shows rare-earth specific Pauli-limit violation in all crystallographic directions [10], which suggests a richer superconducting landscape in the newfound nickelate beyond a cuprate-like image.

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The elusive fermi surface and electrone-like electronic structure of the infinite layer Nickelates

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The recently discovered Ni-based high temperature superconductors have opened up a flurry of activity in attempts to unravel their detailed electronic structure including the Fermi surface. In this paper, we demonstrate that the novel nickelates display some electronic properties that are similar to those of electrone-like materials, thus challenging the Zaanen-Sawatzky-Allen classification paradigm. Oxide based electrone-like materials are materials in which topotactic removal of the most weakly bound oxygens leaves behind voids with a landscape of attractive potentials for electrons. We find that this is also what happens in the infinite-layer compound $RNiO_2$ (R =rare-earth atom) where one of the extra 2 electrons needed for charge neutrality when the apical oxygen is removed from the cubic perovskite $RNiO_3$ is to a large degree located in the oxygen vacancy position rather than mainly used to convert Ni from $3+$ to $1+$. This is shown to be the main reason for the theoretical difficulty in determining the much debated elusive Fermi surface of the infinite-layer nickelates. This effect of the inclusion of states centered at the O vacancy position also strongly modifies the effects of electron correlation as we also demonstrate in an extended impurity like many body exact diagonalization calculation.

Emergent Intrinsic Transport Properties of Infinite-Layer Nickelates

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The discovery of superconductivity in the infinite-layer nickelates [1] has sparked significant interest due to their structural and electronic similarities to the superconducting cuprates [2]. In parallel, the additional hybridized electron bands in the nickelates stand out as a marked difference from the cuprates [3,4], and whether this manifests in the macroscopic physical properties has been a central question. Thus far, initial experimental investigations have revealed important distinctions from the cuprates in the orbital alignment [5,6] and ground states in the superconducting phase diagram of the infinite-layer nickelates [7,8], which have been largely attributed intrinsically to the multiband nature of the nickelates [9]. However, the limited crystallinity of these thermodynamically unstable materials [10] introduces uncertainties to this conclusion, strongly calling for improved materials control in the nickelates to investigate their intrinsic superconducting and normal-state properties.

In continuation of our efforts to optimize the synthesis of the infinite-layer nickelates [10], we have recently achieved an important breakthrough in the materials improvement of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ thin films, with consistently high materials quality with strong suppression of Ruddlesden-Popper-type stacking faults across a wide range of hole doping $x = 0.05\text{-}0.325$. Investigation of the phase diagram on these films reveal intrinsic transport properties of the infinite-layer nickelates, including a correlated ground state in the underdoped region, T -linear strange metal behavior in the optimal doping, and Fermi-liquid ground state in the overdoped region [11]. The details of the emergent intrinsic phase diagram and their implications in relation to the cuprates, as well as the critical aspects of the synthesis optimization leading to high-quality stabilization of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$, are to be discussed.

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Charge density wave and magnetic excitations in infinite layer nickelates

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Superconductivity in infinite-layer nickelates [1] holds exciting analogies with that of cuprates, with similar structures and similar electronic configuration of Cu^{2+} and Ni^{1+} $3d$ states in the CuO_2 and NiO_2 planes, respectively. Yet, they show also distinct properties, like a larger energy separation between the O-2p and the transition metal-3d bands (4eV in nickelates vs. 2eV in cuprates) [2], and an important hybridization between Nd-5d and Ni-3d states which give rise to an electron-like band crossing the Fermi level [2].

Using resonant inelastic x-ray scattering (RIXS) we studied electronic and magnetic excitations and charge density correlations in $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ thin films and we found a surprising dichotomy between the presence of dispersing magnetic excitations and in-plane charge order correlations which depends on the presence/absence of a SrTiO_3 capping layer [3]. The charge order shows up as peak in the elastic resonant scattering in the uncapped $x=0$ compound at wave vector close to $(1/3,0)$ and $(0,1/3)$, remindful of the charge order signal in hole doped cuprates. The peak weakens at $x=0.05$ and disappears in the superconducting $x=0.20$ film. The uncapped samples also present a higher degree of Ni3d-Nd5d hybridization and a smaller anisotropy of the Ni3d occupation with respect to the capped samples, which might explain the observed dichotomy.

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Structural and electronic coupling in transition metal oxide heterostructures

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In this talk, I will illustrate through some examples how electronic and structural coupling can lead to exciting novel properties in oxide films and heterostructures.

I will first recall how tri-linear coupling of structural instabilities can lead to improper ferroelectricity in $\text{PbTiO}_3/\text{SrTiO}_3$ superlattices[1]. I will then discuss how such a coupling of instabilities can be used to design artificial ferroelectric and possibly multiferroic materials with a focus on vanadate-based structures [2,3]. In these structures, it was found that the competition between the strain state and the coupling of oxygen octahedra rotations leads to an interesting “transition layer” and a reorientation of the orthorhombic long axis. This reorientation – that can be used for designing an artificial interface – is also observed in $\text{SmNiO}_3/\text{NdNiO}_3$ nickelate superlattices - in which an electronic coupling of the metal-insulator transition was observed below a critical wavelength [4,5] - when an additional insulating spacer of LaAlO_3 is introduced.

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Controlling the electronic structures of atomically thin oxide films: Hund metals and Mott insulators

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Efforts have been made to manipulate the electronic properties of atomically thin oxide films. Growth and realizing desired properties of atomically thin films are usually done through synthesis of superlattices. As a result, direct observation of the electronic property via, for example, angle resolved photoelectron spectroscopy (ARPES) has been difficult. The difficulty can be overcome by using *in situ* ARPES experiments on atomically thin films. In this presentation, I will present our recent results of *in situ* ARPES studies on transition metal oxide ultra-thin films.

We started with ARPES studies on a few unit cell (uc) thick films of SrRuO₃, a prototypical metallic ferromagnet with spin-orbit coupling. It was found that nodal lines and quadratic band crossing points are generic features of ultrathin perovskite films. These symmetry-protected nodal lines and quadratic band crossing points are sources of Berry curvature that causes the sign changing anomalous Hall effects [1]. By using an additional ‘conducting layer’, we were able to obtain the electronic structure of 1 u.c. thick SrRuO₃ films. Our results show that 1 uc films are not insulators but remain metallic. Doping experiments reveal that 1 uc films are correlated Hund metals caused by the high density of states near E_F from the van Hove singularity [2]. We further controlled the strain and octahedron distortion of 1 uc films by using various substrates. We demonstrate that the electronic state of 1 uc films can be manipulated from a good metal to a correlated Hund metal, and finally to a Mott insulator [3][4].

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Navigating Atomically Precise Synthesis of Stubborn Metal Oxides using Chemistry

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Molecular-beam epitaxy (MBE) has come to the forefront for the thin film synthesis of materials with an exceptionally high structural quality, and with the best figures of merits whether it is related to their electrical or optical properties. From its beginnings as a successful method for III–V semiconductor growth to today for the growth of many contenders for next-generation electronics and spintronics devices, several new synthesis challenges have however emerged. For instance, it has been notoriously difficult to grow metal oxides, in an atomically precise manner, of metals having ultra-low vapor pressures and difficulty of oxidation.

In this talk, we will review these issues and will present our group’s effort to address these challenges using a *novel solid-source metal-organic MBE* approach. Using model material systems based on ruthenates and iridates, we show, for the first time, controlled synthesis of metal and metal oxides of these “stubborn” elements (Ru, Ir, and Pt) with the *same ease and control* as afforded by III-V MBE. For instance, a record low residual resistivity or the residual resistivity ratio was obtained in RuO₂ and SrRuO₃ films, respectively. We will discuss these results in the context of point defect control using adsorption-controlled growth.

Finally, we will present detailed growth study utilizing chemistry of source materials as a controlling knob to navigate synthesis. With the goal to understand and control electronic ground states in defect-managed complex oxide films and nano-membranes, we will discuss how chemistry of source materials can be used to navigate synthesis on-demand [1,2].

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Atomic-scale analysis of the interface between polar and nonpolar LaInO₃/BaSnO₃ perovskite oxides

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The wide-bandgap oxide materials have lately attracted a lot of interest because of the 2-dimensional electron gas (2DEG) at their interface and the high potential for novel electronic devices and applications [1,2]. Particularly, LaInO₃/BaSnO₃ heterostructures were intensively investigated due to the high electron mobility of $\sim 300\text{cm}^2/\text{Vs}$ in BaSnO₃ at room temperature and the formation of a 2DEG at the interface [3]. In case of the prototypical LaAlO₃/SrTiO₃ system, the origin of the 2DEG is attributed to the polar discontinuity and an electronic reconstruction at the n-type LaO-TiO₂ interface [4]. Controlling the interface termination therefore is crucial to accomplish heterostructures with desired properties. In this contribution, we combine the efforts of atomic resolution transmission electron microscopy and density-functional theory, to study the interface between BaSnO₃ and LaInO₃. Access to the atomic structure and composition of the interface is gained by transmission electron microscopy, energy dispersive X-ray spectroscopy, and electron energy loss spectroscopy (EELS). Experiment and theory are in excellent agreement and show that free BaSnO₃ (001) surfaces are BaO terminated, while the interface between BaSnO₃ and LaInO₃ is SnO₂ terminated. This finding indicates that during growth of LaInO₃ layer on BaSnO₃ Ba atoms exchange from the subsurface to the surface. Preliminary EELS analysis of several monolayer thick LaInO₃ layer on BaSnO₃ shows indications of Ba atoms on the LaInO₃ surface in BaSnO₃/LaInO₃ heterostructure, confirming that atomic exchange in this system promotes the energetically favorable SnO₂-LaO interface.

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Exotic magnetic anisotropy near dimensional Mott boundary

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Low-dimensional Mott systems foster intriguing quantum phenomena associated with spins, including electromagnetic coupling, quantum criticality, colossal magnetoresistance, quantum oscillation, Higgs mode, and quantum spin liquid phase. Especially, enhanced magnetic anisotropy in low-dimensional Mott systems was found to be essential in realizing novel electromagnetic phases and resultant magneto-transport behaviors, in materials such as CrI₃, VI₃, Ca₃Ru₂O₇, and Sr₂IrO₄. Most of these natural layered materials, however, have limitations in actively manipulating the dimensionality and/or degree of magnetic anisotropy since the anisotropy is inherently determined by the low-dimensional crystal structure.

Here, we present an intriguing modulation of magnetic anisotropy near a dimensional Mott boundary, stemming from the anisotropic control of the exchange interaction in synthetic oxide heterostructures. Particularly, we built superlattices composed of magnetic monolayer SrRuO₃ and a few layers of nonmagnetic insulating SrTiO₃, i.e., [1|y] superlattices [1-9]. In those heterostructures, the interlayer coupling strength between the SrRuO₃ layers was digitally controlled as a function of the SrTiO₃ unit cell thickness. This leads to the desired dimensional Mott transition coupled to the magnetic transition in the [1|y] superlattices. Especially, when the interlayer coupling strength was maximized at $y = 1$, we observed a largely tunable angular magnetoresistance of SrRuO₃ monolayer superlattice depending on the strength of the external magnetic field and temperature. Density functional calculations implied that the energy difference between in-plane and out-of-plane spin states becomes nearly degenerated for the [1|1] superlattice owing to their quasi-two-dimensional spin order, which would be highly susceptible to both the thermal and magnetic energy scales. The highly unprecedented tuning of the magnetic anisotropy via external magnetic field envisions novel spintronic devices exploiting the spin ordering in low-dimensional Mott systems.

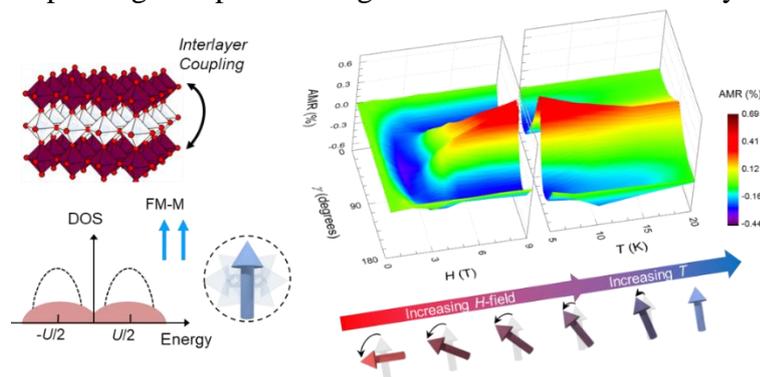


Figure 1. (left) Schematic of dimensional Mott transition in atomically controlled [1|y] superlattices. (right) Contour plot of angular magnetoresistance for [1|1] superlattice as functions of external magnetic field and temperature. The direction of the arrows indicates the magnetic easy axis of the system.

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Magnetotransport Properties of Epitaxial Sr₃Ru₂O₇ -based Heterostructures and Devices

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The system Sr₃Ru₂O₇ belongs to the strontium ruthenate oxides of the Ruddlesden Popper series, Sr_{n+1}Ru_nO_{3n+1} ($n = 1, 2, 3, \infty$) [1], with $n = 2$. In contrast to Sr₂RuO₄ ($n=1$), which is an unconventional superconductor [2, 3]; Sr₄Ru₃O₁₀ ($n=3$), which exhibits anisotropic ferromagnetism and orbital-dependent metamagnetism [4,5] and SrRuO₃ ($n=\infty$), which presents spontaneous itinerant ferromagnetism [6]; Sr₃Ru₂O₇ is a paramagnetic metal in its ground state [7,8]. The material Sr₃Ru₂O₇ exhibits a range of unusual emergent properties, such as quantum critical metamagnetism and electron nematic behavior [7,8]. These unusual ground state properties make Sr₃Ru₂O₇ a good playground to explore novel electronically or magnetically ordered quantum phases and their relationship to the underlying band structure, as well as diverse oxide-based electronic device directions.

Only a few attempts have been reported for the growth of Sr₃Ru₂O₇ thin films [9,10], and little is known about the magnetic ground state and magnetotransport properties of epitaxial Sr₃Ru₂O₇ films and quantum devices. Using pulsed laser deposition (PLD), we have prepared Sr₃Ru₂O₇-based heterostructures from a stoichiometric Sr₃Ru₂O₇ target and a RuO₂ target that were ablated alternately (the RuO₂ target was used to compensate for Ru loss during PLD growth). We report systematic magnetotransport measurements of Sr₃Ru₂O₇-based heterostructures and sub-micrometer-size Hall bar devices fabricated from such heterostructures using electron beam lithography. The origin of the observed large magnetoresistance peak potentially induced by the metamagnetic transition in Sr₃Ru₂O₇ will be discussed. These results open a new route for the precise exploration of size effects in the magnetic properties of Sr_{n+1}Ru_nO_{3n+1} compounds by using ultra-thin quantum devices.

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Charged Higher Order Topologies in Room Temperature Magnetoelectric Multiferroic Thin Film

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Magnetoelectric multiferroic higher order topologies are an emerging solution for future low-power spintronic and quantum devices, due to a combination of their atomic-scale tunable multifunctionality and mobility. In addition to their potential for next-generation interactive technology, topologies with spin to charge coupling are a fertile ground for the exploration and engineering of new states of matter. In our recent report we have shown the presence of vortex topologies present in room temperature magnetoelectric multiferroic $\text{Bi}_6\text{Ti}_x\text{Fe}_y\text{Mn}_z\text{O}_{18}$ thin films by atomic-scale scanning transmission electron microscopy (STEM) polarization mapping.[1,2] We find these chiral polar textures at antiphase boundaries within the thin films. Such topologies related to crystalline defects should be analyzed three-dimensionally to truly understand their formation. In this presentation, will discuss the use of multi-dimensional microscopy techniques such as atom probe tomography (APT) combined with scanning transmission electron microscopy (STEM) and spectroscopies to analyze the Fe and Mn elemental segregation at topological defects.

For $\text{Bi}_6\text{Ti}_x\text{Fe}_y\text{Mn}_z\text{O}_{18}$ the magnetic cation partitioning increases the probability of nearest-neighbor magnetic interactions in the central unit cell layer by up to 90% compared to random distributions over the other available B-sites.[3] Using atomic resolution STEM with corresponding energy dispersive X-ray (EDX) and electron energy loss spectroscopy (EELS) elemental mapping we can determine the location of Fe and Mn within the unit cell. Although electron spectroscopy characterization is also possible for topological defects, using this method we only have a 2D projected view of these 3D microstructural features. As the topologies themselves are the active regions within such material systems, it is vital that we explore the higher order elemental segregation in full real space. We hence used APT's unique 3D characterization capability to move beyond the 2D projection of the elemental segregation knowledge, with an aim to unravel the magnetic cation partitioning of complex 3D vortices and associated crystallographic defect topologies. Theoretical calculations confirm the subunit cell cation site preference and charged topology energetics. We will discuss the opportunities APT characterization methods can bring to the field multiferroic higher order topology physics and other oxide electronics research.

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How defects and dopants stabilize ferroelectricity in HfO₂ – a spectroscopic approach

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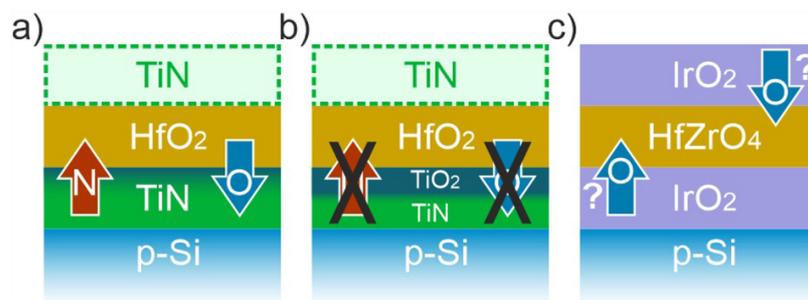
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HfO₂-based ultrathin films are key multifunctional material class for nonvolatile ferroelectric field effect transistors, memristors or novel neuromorphic computing concepts. Oxygen vacancies and metallic impurity ions are recognized as defects which both can stabilize the ferroelectric phase in thin film HfO₂.

Hard X-ray photoelectron spectroscopy (HAXPES) provides a unique tool to explore the interface properties and tunability of the oxygen content in HfO₂-based devices. We present studies on the emerging electronic states of HfO₂-based capacitors and focus on the identification of the (mostly indirect) signatures of oxygen vacancies or rare-earth dopants in the HfO₂ layers and at the metal electrode interfaces [1].

We correlate HAXPES data recorded at the beamline P22 (DESY Hamburg, Germany) to lab-based electric field cycling experiments, and conclude on how the microscopic HfO₂ electronic structure directly relates to macroscopic ferroelectric polarization and eswitching properties. We show selected examples which reveal the different interface oxygen exchange mechanisms in HfO₂-metal electrode structures [2], demonstrate the oxygen vacancy formation in HfO₂ layers dependent on rapid thermal annealing processes [3], as well as oxygen vacancy/nitrogen impurity interactions altered by deposition-specific parameters [4].



Oxygen exchange mechanisms in metal-insulator-metal capacitors of ferroelectric HfO₂ and metallic electrodes: Dependent on the choice of the metal electrodes, ferroelectric functionality is altered by (a) oxygen scavenging by a TiN electrode, (b) interface passivation by a TiO₂ interlayer, and (c) oxygen supply by IrO₂ electrodes.

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Negative Capacitance Electronics via Ultrathin Ferroelectricity in HfO₂-ZrO₂

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The two-dimensional (2D) limit of ferroelectricity, an active topic in condensed matter research, also has extensive technological implications for the scaling of energy-efficient electronics. In particular, the negative capacitance (NC) effect in ferroelectrics has emerged to overcome fundamental computing energy limits. However, advanced semiconductor technology integration requires NC stabilization at the ultrathin regime on silicon, posing significant materials challenges for conventional ferroelectrics.

To address this, we first established ultrathin ferroelectric order down to 1-nm and 5-Å thickness in doped HfO₂ [1,2] and conventionally antiferroelectric ZrO₂ [3], respectively, the same high-κ dielectrics used in today's advanced logic and memory technologies. These binary oxide films on silicon boast ultrathin-enhanced polarization signatures [1-3] – in stark contrast to conventional perovskite oxide ferroelectrics – marking a breakthrough towards exploiting ferroic-based phenomena at ultra-scaled dimensions for electronics.

Next, we leveraged the competing atomic-scale (anti)ferroelectric orders to design NC in sub-2-nm HfO₂-ZrO₂ superlattices [4] and 1-nm ZrO₂ [5], the gate oxide thicknesses required for today's state-of-the-art transistors and future 3D architectures, respectively. Capacitance enhancement via NC has been demonstrated for many perovskite-structure ferroelectric–dielectric systems; this work is the first demonstration that the same enhancement is possible in HfO₂-ZrO₂ fluorite-structure system. Furthermore, this seamless materials swap – conventional high-κ dielectric HfO₂-ZrO₂ to (anti)ferroelectric HfO₂-ZrO₂ – demonstrates key performance benefits in NC transistors compared to established semiconductor foundry benchmarks [4,5].

Therefore, leveraging these materials breakthroughs at the 2D limit – ferroelectricity [1-3] and negative capacitance [4-5] – provides a realistic pathway towards energy-efficient electronics via integrated ferroelectrics. And from a thin film perspective, this approach to exploit 3D materials confined to the 2D limit – particularly within this simple binary oxide model system hosting unconventional and emergent ferroelectric size effects [1-3] – offers an atomic-scale approach to unlock previously hidden electronic phenomena [3-5].

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Ferroelectrics Everywhere

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Ferroelectricity in wurtzite-based crystals was observed in 2019 and immediately introduced exciting opportunities to explore and discover new structure-property relationships in novel formulation spaces, and to investigate new integration and device implementations given new process compatibilities. The seminal discovery of ferroelectric $\text{Al}_{1-x}\text{Sc}_x\text{N}$ by Fichtner *et al.*¹ initiated this excitement and was followed by comparable observations of polarization reversal in the structurally similar $\text{Al}_{1-x}\text{B}_x\text{N}$ ² and the $\text{Zn}_{1-x}\text{Mg}_x\text{O}$ ³ systems. These observations lead one to speculate that ferroelectricity might be found much more broadly, even “everywhere”, by introducing the appropriate disorder.

In this presentation the structure-process-property relationships in the B-substituted AlN and Mg-substituted ZnO wurtzite systems will be demonstrated. The B-substituted materials exhibit square hysteresis loops with polarization values between $150 \mu\text{C}/\text{cm}^2$ and $120 \mu\text{C}/\text{cm}^2$ when boron concentrations range between 2% and 15% respectively. Coercive field values fall with additional boron, from $5.5 \text{ MV}/\text{cm}$ to about $5 \text{ MV}/\text{cm}$ at B saturation. Bandgap values are approximately 5 eV or above in all cases. Material can be prepared between 100°C and 350°C with very little difference in electrical properties. W bottom and top electrodes are used in all cases. Capacitors can be prepared down to 50 nm thick before leakage current becomes problematic during low frequency hysteresis measurements. First principles calculations that rationalize the unit cell volume, bond angle distribution, and remanent polarization will be presented.

Comparable results are found in the $\text{Zn}_{1-x}\text{Mg}_x\text{O}$ system. Between 25% and 35% Mg substitution, square hysteresis loops with remanent polarization values above $100 \mu\text{C}/\text{cm}^2$ are readily achieved. Transmission measurements show bandgap values between 4.0 eV and 4.2 eV in this range. In comparison to AlBN, coercive field values for ZMO are as low as $1.7 \text{ MV}/\text{cm}$.

In addition to a discussion of properties and preferred synthesis routes, we will discuss the limitations to property engineering and scaling, and provide a complete update on observations/explorations in additional compositional and structural families.

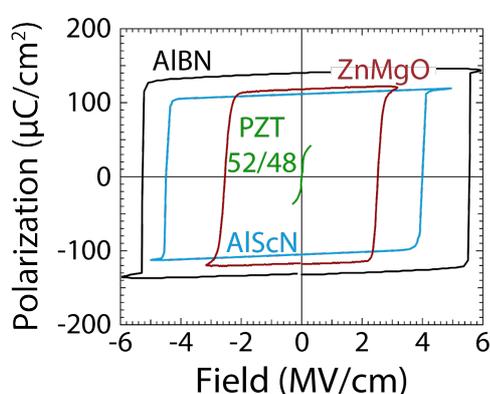


Figure 1: Polarization hysteresis loops for AlBn, AlScN, AlBN and PZT thin film capacitors. This illustrates the property contrast, i.e., polarization and coercive field, between perovskite and wurtzite ferroelectric materials.

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Signatures of enhanced out-of-plane polarization in asymmetric BaTiO₃ superlattices integrated on silicon

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To bring the diverse functionalities of transition metal oxides into modern electronics, it is imperative to integrate oxide films with controllable properties onto the silicon platform. Here, we present asymmetric LaMnO₃/BaTiO₃/SrTiO₃ superlattices fabricated on silicon with layer thickness control at the unit-cell level [1]. By harnessing the coherent strain between the constituent layers, we overcome the biaxial thermal tension from silicon and stabilize c-axis oriented BaTiO₃ layers with substantially enhanced tetragonality, as revealed by atomically resolved scanning transmission electron microscopy. Optical second harmonic generation measurements signify a predominant out-of-plane polarized state with strongly enhanced net polarization in the tricolor superlattices, as compared to the BaTiO₃ single film and conventional BaTiO₃/SrTiO₃ superlattice grown on silicon. Meanwhile, this coherent strain in turn suppresses the magnetism of LaMnO₃ as the thickness of BaTiO₃ increases. Our study raises the prospect of designing artificial oxide superlattices on silicon with tailored functionalities.

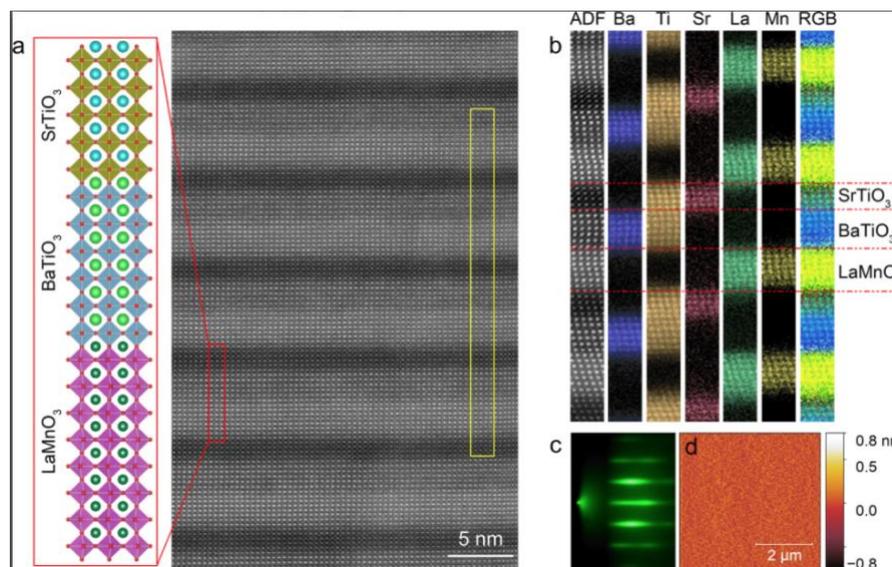


Fig. 1 LMO/BTO/STO SLs on Si/STO. a HAADF-STEM image of the L8B6S5 SL grown on Si/STO. The sketch on the left shows the ideal atomic structure in one period. The EELS elemental maps taken from the yellow rectangular area are shown in (b). c, d Show the RHEED image taken along the Si[110] azimuth and AFM image of the L8B6S5

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Pairing Mediated by the Transverse Optic Mode in KTaO_3

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Recently, superconductivity has been discovered for the electron gas formed at the interface of KTaO_3 with other oxides [1], T_c being almost an order of magnitude larger than what is seen for SrTiO_3 . The largest T_c is for $\langle 111 \rangle$ interfaces, with T_c for $\langle 110 \rangle$ interfaces about half of this, and that for $\langle 001 \rangle$ interfaces either extremely small or non-existent. This points to the role of orbital degeneracy, the degeneracy of t_{2g} states being maximal for $\langle 111 \rangle$, and completely lifted for $\langle 001 \rangle$ due to interface confinement. A theory that encompasses this effect is pairing via the transverse optic mode that leads to ferroelectricity in related perovskites, as to linear order this involves inter-orbital interactions (which in turn gives rise to a large Rashba effect that is strongly sensitive to orientation). In this talk, I will compare this theory to relevant data for KTaO_3 , in particular the doping dependence of T_c and how T_c and thermodynamic and transport properties vary with interface orientation [2].

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Interface band engineering in LaAlO₃/SrTiO₃ heterostructures

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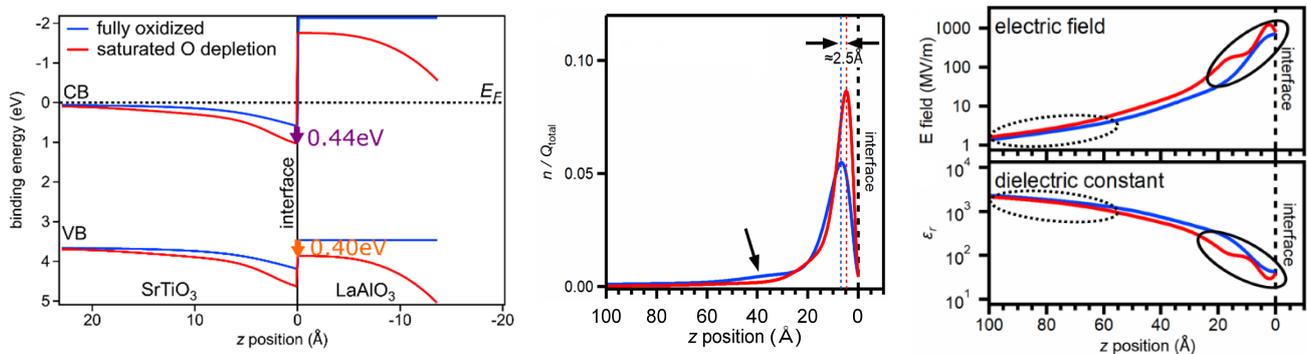
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Novel two-dimensional electron systems at the interfaces of oxide heterostructures such as LaAlO₃/SrTiO₃ have attracted much attention as they open a new route to harness the rich quantum phases of transition-metal oxides for potentially useful functionalities not available in conventional semiconductor electronics. For such applications the controllability of the interface properties such as band bending and offset is key.

We demonstrate that these properties can be effectively engineered in LaAlO₃/SrTiO₃ by virtue of the oxygen vacancy concentration. By analyzing angle-dependent hard x-ray photoelectron spectroscopy (HAXPES) data with a Poisson-Schrödinger model, we derive a complete band diagram and charge-carrier profile of the heterostructure dependent on the oxygen vacancy concentration which is systematically controlled during the photoemission experiments by means of synchrotron light irradiation and simultaneous oxygen dosing. We observe a crossover of the band alignment from type II to type I with increasing doping level, which reconciles the striking inconsistencies among previous photoemission studies. In particular, the dielectric constant of the SrTiO₃ substrates and its strong non-linear dependence on the electric field prove to be essential for the band arrangement of the LaAlO₃/SrTiO₃ heterostructure.



Band scheme and electrostatics at the (001)-oriented LaAlO₃/SrTiO₃ heterostructure in the fully oxidized and an strongly oxygen depleted state.

Moiré engineering and novel electronic transport at oxide interfaces

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Moiré engineering has recently emerged as an effective approach to control quantum phenomena in condensed matter systems. Here we demonstrate electronic moiré patterns in films of a prototypical magnetoresistive oxide, $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$, epitaxially grown on LaAlO_3 substrates. Microscopic moiré profiles of both the electronic conductivity and ferromagnetism are observed, which can be attributed to the coexistence and interaction of two distinct incommensurate patterns of strain modulation. Our work provides a potential route to achieving spatially patterned electronic textures on demand in strained epitaxial materials [1].

On the other hand, the states inside superconducting gaps in tunneling spectra have been revealed to be associated with a spectrum of nontrivial effects, such as Majorana bound states. Here we further present our recent progresses in the tunneling measurements involving oxide heterostructures, and the central findings include: 1) Zero-bias conductance peaks effectively tuned by gating-controlled Rashba spin-orbit coupling in the hybrid junctions composed of the conducting $\text{LaAlO}_3/\text{SrTiO}_3$ interface and superconducting Nb [2]; 2) In-gap Andreev bound states observed for the hybrid junctions involving the newly discovered superconducting $\text{LaAlO}_3/\text{KTaO}_3$ interface, which can be attributed to unconventional superconductivity [3]. These discoveries collectively have implications in various research fronts from superconducting spintronics to topological superconductivity.

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Superconducting epitaxial thin films of (111) KTaO_3 grown by hybrid PLD

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Modern quantum materials are inherently sensitive to point defects, and require a new synthesis route to produce epitaxial oxide thin films and interfaces clean enough to probe fundamental quantum phenomena. The recent discovery of robust superconductivity at KTaO_3 (111) and KTaO_3 (110) heterointerfaces on KTaO_3 bulk single crystals offers new insights into the role of incipient ferroelectricity, strong spin-orbit coupling, and structural phase transitions. Electronic grade epitaxial thin film platforms will facilitate investigation and control of the interfacial superconductivity and understanding the fundamental mechanisms of the superconductivity in KTaO_3 . The major challenge of research on KTaO_3 system is that it is difficult to grow high-quality KTaO_3 epitaxial thin films due to high volatility of potassium. Recently, we have developed the hybrid PLD method for electronic grade KTaO_3 thin film growth, which successfully achieves this by taking advantage of the unique capabilities of PLD to instantly evaporate Ta_2O_5 in a controlled manner and evaporation of K_2O to maintain sufficient overpressure of volatile species. We successfully synthesized heteroepitaxial KTaO_3 thin films on (111)-oriented KTaO_3 bulk single crystal substrates with a SmScO_3 template by hybrid PLD, followed by a LaAlO_3 overlayer. AFM and TEM data show that the KTaO_3 thin films are epitaxial to the bulk single crystal KTaO_3 (111) substrates with atomically flat surfaces. Electrical transport data show a superconducting transition temperature of ~ 1 K. We anticipate that the ability to synthesize high-quality epitaxial complex oxides such as KTaO_3 that contain volatile elements will provide a new platform for exploring new physics and technological applications arising from unique characteristics such as large spin-orbit coupling.

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Interface and surface superconductivity in KTaO_3

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A new family of oxide-interface superconductors, with a transition temperature (T_c) up to 2.2 K, were recently found at interfaces between KTaO_3 and other oxides (EuO or LaAlO_3) [1-3]. In this talk, we will present our recent progresses in this system. We confirm that the superconductivity depends strongly on the orientation of KTaO_3 and find a $T_c \sim 0.9$ K superconductivity at (110)-orientated interface [2]. We control the superconductivity of (111)-orientated interface with electric field, achieving a superconductor-to-insulator quantum phase transition and observing a tunable quantum metal behavior [3]. We explain the tuning effect by change in the spatial profile of the carriers near the interface and hence, effective disorder, rather than carrier density. By in-situ transport measurements during growth, we conclude that the formation mechanism of the $\text{LaAlO}_3/\text{KTaO}_3$ interface electron gas is electron transfer from oxygen vacancies in LaAlO_3 film to KTaO_3 substrate [4]. By exploiting ionic liquid gating, we obtain superconductivity at bare $\text{KTaO}_3(111)$ and (110) surfaces with T_c value up to 2.0 K and 1.0 K, respectively [5]. This oxide-interface-free superconductivity provides a clear evidence that the essential physics of KTaO_3 interface superconductivity lies in the KTaO_3 surfaces doped with electrons. We also demonstrate a non-volatile control of $\text{LaAlO}_3/\text{KTaO}_3(111)$ superconductivity by surface charges writings using a biased conducting atomic force microscopy tip [6].

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Transient electron scavengers modulate carrier density at a polar/nonpolar perovskite oxide heterojunction

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Oxide heterostructures that facilitate the formation of two-dimensional (2D) electron gases are promising candidates for novel classes of electronic devices. We show how transient electron scavengers can be utilized to control the 2D carrier concentration (n_{2D}) at polar/non-polar perovskite interfaces. By combining quantitative synchrotron x-ray-based interface structure determination (COBRA) with *ab initio* modeling, we demonstrate [1] that Nd vacancy (V_{Nd}) formation and the resulting formation of Nd adatoms, stabilized by oxygen scavengers at the growth front, can quantitatively account for the decreased n_{2D} at the SrTiO₃ / n NdTiO₃ / SrTiO₃ (001) heterojunction for $n=1$ [2]. The proposed mechanism stems from the ability of the oxygen species adsorbed at the surface to trap electron charge and, therefore, create near-surface electric field. This effect is independent of the synthesis approach if (i) electron scavengers are present at the growth front and (ii) the material in question is ionic, i.e., the near-surface electric field is compensated by rearrangements of the neighboring ionic species. The mechanism applies to any NTO thickness, and explains how the accumulation of V_{Nd} and displaced Nd results in the apparent Sr-Nd intermixing reported at the STO/NTO interfaces [3]. These new insights into growth mechanisms and the effect of transient species and defects on the electronic properties of oxide heterojunctions suggest that utilizing electron scavengers that are more aggressive than oxygen, or displacing electron scavengers with inert gases, could be used to control vacancies of A-site species and, therefore, the n_{2D} .

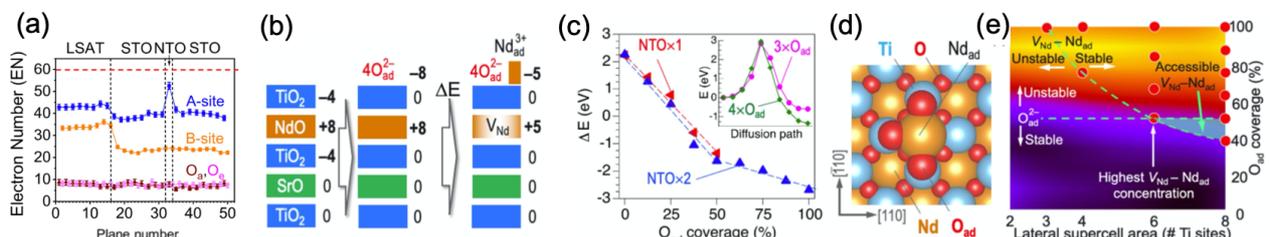


Figure caption: The existence of Nd vacancies in the NdO plane [1] evident from COBRA measurements (a) is explained by the ability of O/O₂ electron scavengers to bind at the NTO surface (b); the resulting electron field promotes the formation of Nd vacancy–adatom defect pairs (c) stabilized by adsorbed oxygens (d). Estimated concentration of these defects explains the observed decrease of the n_{2D} [3].

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Anisotropic 2D Superconductivity at KTaO_3 (111) Interfaces

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Two-dimensional (2D) superconductivity is a fertile landscape for novel and exotic quantum phenomena. Earlier studies were focused on understanding the quantum phase transitions (QPTs) in 2D disordered systems. Recently, highly crystalline 2D superconductors have allowed observation of intrinsic quantum phenomena, such as violation of paramagnetic Pauli limit, quantum metallic state, and quantum Griffiths phase. Spin-orbit coupling (SOC) in crystalline 2D superconductors leads to a robust superconducting state, surviving anomalous applied magnetic fields.

An anisotropic 2D superconductivity was recently discovered in KTaO_3 interfaces [1]. The superconducting order parameter is sensitive to the interface orientation with critical superconducting temperatures of 0.05 K, 1 K, and 2 K for (100), (110), and (111) interfaces. Furthermore, high quality KTaO_3 (111) interfaces show an anisotropic superconducting transition along [110] and [112] directions. The spin splitting and magnitude of spin-orbit coupling is highly sensitive to the crystallographic direction [2].

We grew high quality metallic KTaO_3 interfaces, using an oxide molecular beam epitaxy (MBE). The cross-section high-angle annular dark-field (HAADF) imaging in scanning transmission electron microscopy (STEM) shows an atomically abrupt interface without any extended defects. The sheet resistance v.s. temperature demonstrates a metallic behavior, $dR/dT > 0$, extending to 2 K. The Hall carrier density is $\sim 6 \times 10^{13} \text{ cm}^{-2}$ at 2 K, resolving a carrier mobility of $\sim 200 \text{ cm}^2/\text{v.s.}$ We observe an anisotropic superconductivity emerges below 1.4 K. The in-plane critical field (H_{c2}) exceeds the Pauli limit and is sensitive to bias and magnetic field directions. Figure 1 shows the in-plane H_{c2} along $\langle 110 \rangle$ and $\langle 112 \rangle$ bias directions. We observe anisotropic behavior in both critical temperature and critical field at the KTaO_3 interfaces. Interestingly, the H_{c2} depends on both bias and applied field directions.

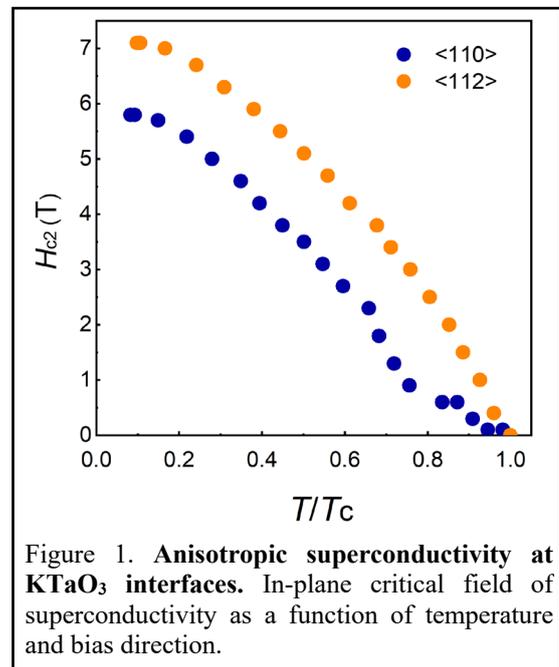


Figure 1. **Anisotropic superconductivity at KTaO_3 interfaces.** In-plane critical field of superconductivity as a function of temperature and bias direction.

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Electrochemical Modulation of Electronic States in Strongly Correlated Transition-Metal Oxides

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In order to induce various electronic phase transitions in strongly correlated electron systems, lithium-ion battery-like electrochemical cells based on highly crystalline epitaxial thin films of transition-metal oxides (TMOs) have been developed (Fig. 1). The electronic states of band insulators (MoO_3 [1] and WO_3 [2]), Mott insulators (LiRhO_2), heavy fermion metals (LiV_2O_4 [3]), and superconductors (LiTi_2O_4 [4], Cs_xWO_3 [5], LiNbO_2 [6,7]) are studied in detail from their transport, optical, and structural properties. This method has become a powerful tool for precise electrochemical doping of single samples and *in-situ* measurement of modulated electronic states (Fig. 2). In this talk, we discuss the results obtained in our previous studies, clarify the effectiveness and scope of application of this method, and look forward to its possible application to electrochemical devices.

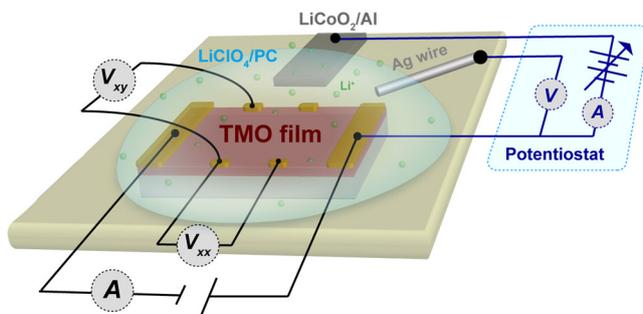


Fig. 1 Schematic illustration of electrochemical Li-ion devices used in this study.

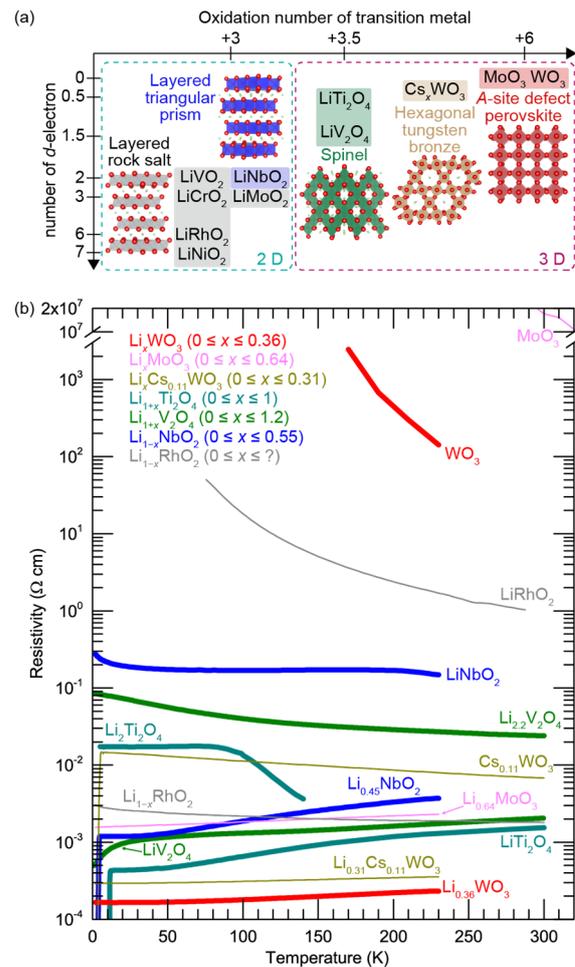


Fig. 2 TMOs investigated in this study. (a) Schematic illustration of the crystal structures. (b) Temperature dependence of the resistivity.

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Orbital Occupancy Inversion in Strained SrVO₃ by Soft-XAS: Surface versus Bulk

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SrVO₃ (SVO) is a d^1 correlated metal with a cubic perovskite structure. It presents a metal to insulator transition as part of the filling system La_{1-x}Sr_xVO₃ [1], and recently it gained interest as a promising earth-abundant transparent conductive oxide [2]. Therefore, tuning the SVO physical properties is appealing for future electronic and optoelectronic devices. To that end, we employ epitaxial strain engineering of SVO films by using different substrates to apply tensile and compressive strain. The SVO synthesis is done via oxide molecular beam epitaxy, and we employ structural and electronic characterization to confirm the high quality of the films. While the unstrained cubic SVO has a threefold degenerated V $3d t_{2g}$ atomic orbitals, the symmetry breaking under biaxial epitaxial strain results in preferred orbital occupation. We employ soft X-ray absorption spectroscopy (XAS) in total electron yield (TEY) and X-ray excited optical luminescence (XEOL) modes for surface and bulk measurements, respectively. Using these two modes we derive the X-ray linear dichroism (XLD), which is where the orbital occupancy is manifested. The surface-sensitive XLD spectra don't show dependence on the strain state, which we ascribe to the near-surface region of SVO [3]. However, when employing bulk-sensitive XLD analysis, we observe a change of sign between compressive to tensile strain. This is the first direct evidence of the predicted inverse preferred occupation in this system [4]. These results shed light on the electronic structure under epitaxial strain and question the use of surface-sensitive methods for bulk characterization.

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Electron doping and epitaxial strain effect in quantum well of SrVO₃ grown by gas source molecular beam epitaxy

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Perovskite SrVO₃ with a 3d¹ electronic configuration is a typical strongly correlated electron system. Interestingly, ultra-thin SrVO₃ films are known to be a Mott insulator [1, 2]. In addition, the high crystalline quality films grown by gas source molecular beam epitaxy (MBE) with a metal organic precursor show very low residual resistivity of the order of 10⁻⁷ Ωcm [3]. Such a low residual resistivity seems to reflect the low concentration of impurities and defects in the films. Thus, it is expected that the gas source MBE SrVO₃ films can be a ‘clean’ platform of Mott physics. Here, we investigate the electron doping and epitaxial strain effect for the 3d¹ electron confined structure of SrVO₃ by substitution La for Sr.

For the electron doping into the 2D Mott insulating SrVO₃, Sr_{1-x}La_xVO₃ with various x were grown in the quantum well structure with various thicknesses sandwiched by 5 unit cells (u.c.) thick capping and buffer SrTiO₃ (STO) barrier layers. The lattice constant of cubic SrVO₃ is 3.842 Å. We chose LSAT (a = 3.868 Å) and STO (a = 3.905 Å) as substrates. The lattice mismatch of SrVO₃ layer against each substrate is +0.68% on LSAT and +1.64% on STO, resulting in smaller tensile strained SrVO₃ on LSAT and larger one on STO as schematics of VO₆ octahedron in the Fig. 1. All the samples were epitaxially grown on LSAT and STO(001) substrates in a gas source MBE equipment. As previously reported films, ultra-thin SrVO₃ layers on both substrates show insulating behaviors [1, 2]. By electron doping in such quantum wells, we found a distinct effect of epitaxial strain. In the case of 3 u.c. thickness of Sr_{1-x}La_xVO₃ on LSAT substrate, the resistivity is successfully reduced by the La substitution up to x = 0.17, implying the existence of metallic state as denoted with shaded region, and then returns to insulating state by the additional doping. In contrast, the resistivity for the films on STO almost monotonically increases by the doping as shown in Fig. 1 (c) and (d). These results indicate an unprecedented phase diagram for the two dimensional system of SrVO₃ that will be discussed in more detail in this presentation. These new findings suggest that this artificial SrVO₃ based heterostructure is an ideal 2D Mott insulator system to investigate novel correlated transport phenomena.

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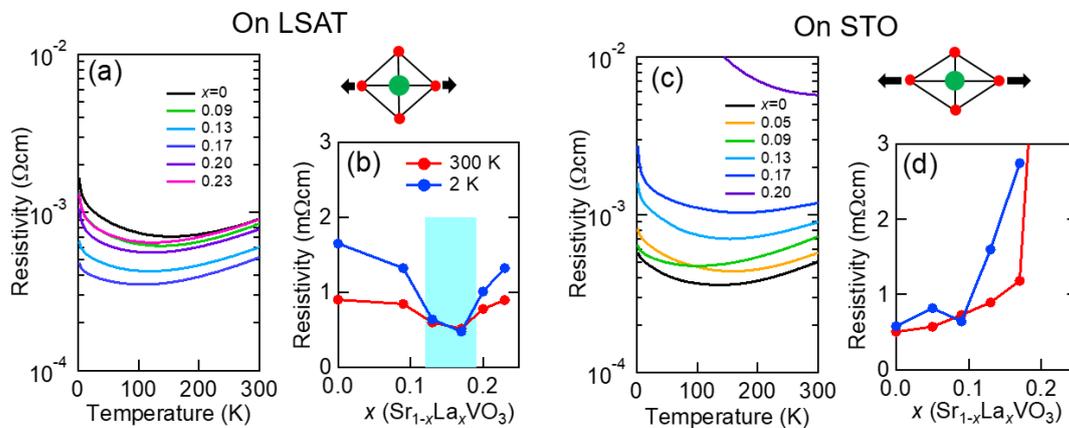


Figure 1: Transport properties of STO(5 u.c.) / Sr_{1-x}La_xVO₃(3 u.c.) / STO(5 u.c.) heterostructures. Temperature dependence of resistivity for the films on LSAT (a) and STO (c). La doping dependence of the resistivity at 300 K and 2 K for the films on LSAT (b) and STO (d).

Strained SrNbO₃: A Correlated Oxide Dirac Semimetal that enters the Extreme Quantum Limit

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Quantum materials (QMs) with strong correlation and nontrivial topology are indispensable to next-generation information and computing technologies. The exploitation of topological band structure is an ideal starting point to realize correlated topological QMs. Among many classes of QMs, complex oxides offer a rich playground to exploit both correlations of Coulombic interactions between electrons often found in 3d transition metal oxides (TMOs) and SOC from heavy metal-based 5d TMOs, such as SrIrO₃ and Na₂IrO₃, leading to many intriguing physical properties. Hybrid materials with proper balancing of the correlation and SOC offer unique opportunities to discover new properties, and heterostructuring or combining dissimilar materials is a great approach to harnessing the complex interplay between quantum wavefunctions and various control parameters, such as proximity, dimensionality, topology, and symmetry. Here, we report that strain-induced symmetry modification in correlated oxide SrNbO₃ thin films creates an emerging topological band structure. Dirac electrons in strained SrNbO₃ films reveal ultrahigh mobility ($\mu_{\text{max}} \approx 100,000 \text{ cm}^2/\text{Vs}$), exceptionally small effective mass ($m^* \sim 0.04m_e$), and nonzero Berry phase. Strained SrNbO₃ films reach the extreme quantum limit (XQL), exhibiting a sign of fractional occupation of Landau levels and giant mass enhancement [1]. Our results suggest that symmetry-modified SrNbO₃ is a rare example of correlated oxide Dirac semimetals, in which the strong correlation of Dirac electrons leads to the realization of a novel correlated topological QM. While this is the second example of oxide-based topological quantum materials after the discovery of CaIrO₃ [2], we note that, when strained, much greater transport properties are discovered with the fractional occupation of Landau levels as it enters XQL at a low magnetic field. *This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

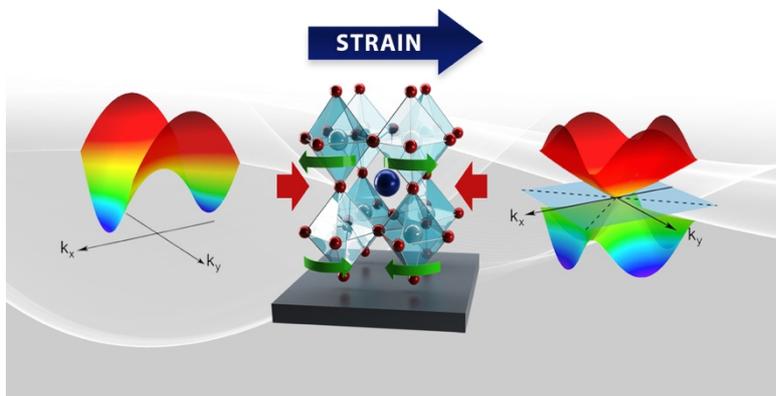


Fig. 1. Schematic representation of a strain-induced Dirac semimetal state in SrNbO₃ that enters XQL with high carrier mobility and linear magnetoresistance.

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Dynamic control of octahedra rotation in perovskites by defect engineering

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Controlling rotations and tiltings of BO_6 octahedra in ABO_3 perovskites is valuable for designing and enhancing functionalities in these materials. However, tuning these degrees of freedom dynamically is challenging, since octahedra rotations do not usually break space inversion symmetry and are, thus, hard to couple with external electric fields.

In this work, we propose a strategy that exploits antisite defects that create local electric dipoles and interact with rotational degrees of freedom [1]. This approach, which relies on substituting an A site with a smaller ion, paves a way to couple dynamically octahedra rotations to external electric fields. Through density functional theory (DFT) calculations we study Al_{La} antisite defect in LaAlO_3 (LAO) as a prototype to demonstrate this concept. We show that the antisite structure is strongly coupled to the surrounding O atoms and by tuning the orientation of the local dipole created by the defect, the nearby octahedra tilts can be modified. We calculate the energetics of possible switching paths, and address the spatial modulation of the local rotational degrees of freedom to the whole lattice.

We note that anti-site defects where the A site is substituted by smaller ions are reported to be widely existing in other systems. Therefore, our strategy would suit not only for LAO but also for other perovskites of similar structure.

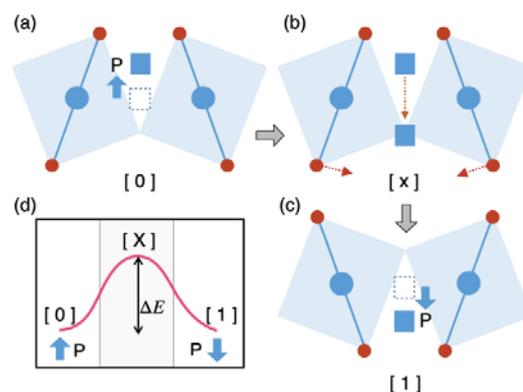


Figure 1. A scheme of the proposed mechanism to control octahedra tilts. The antisite defect (Al substituting La in LAO) moves preferentially towards a position close to the surrounding O atoms, giving rise to a local polarization (a). This polarization can couple to external electric fields, enabling the displacement of the defect to a different configuration (b). The new defect configuration is metastable, and the strong coupling to nearby O atoms modifies the rotation angle of the host lattice octahedra (c). The switching energy barrier (d) has been computed with the NEB method to be 0.33 eV, small enough to make its experimental realization possible

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Interplay Between Structure, Charge, and Spin in Entropy-Stabilized Oxides for Widely Tunable Responses

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Entropy-stabilized oxides (ESOs) demonstrate a new and unprecedented degree of chemical control in materials. Entropy stabilization can be used to incorporate cationic species in an atypical coordination, such as Cu²⁺ and Zn²⁺ into octahedral coordination, and increase the solubility of elements [1,2]. This broadens the compositional space of crystalline oxides and presents opportunities to understand and explore the local chemical and structural disorder resulting from different atomic sizes and the preferred coordination of the constituents. Particularly in oxides, where the physical behavior is strongly correlated to stereochemistry and electronic structure, entropic stabilization creates a unique platform to tailor the interplay between structural and chemical disorder to realize unprecedented functionalities [3]. Here, I will discuss the control of stereochemically-driven structural disorder in single crystalline, rocksalt, (MgCoNiCuZn)O-type entropy-stabilized oxides through the incorporation of Co and Cu cations [4-6]. We harness the disorder to tune the degree of glassiness in the anisotropic antiferromagnetic magnetic structure. Structural distortions driven by the Jahn-Teller effect drive a difference in valence on the Co cation sites, which extends to dilution and disorder of the magnetic lattice. A spin glass model reveals that the fractional spin ordering of the magnetic lattice can be tuned by ~65%. Incorporation of other Jahn-Teller elements show an anomalous (non-Vegard's) behavior in the evolution of the lattice parameter and underlying magnetic transition temperature. After realizing single crystalline films on an epitaxial electrode, our transport measurements reveal a widely tunable conductivity via the Mg concentration that is mediated by hopping and correlated to the global disorder and defect density [7,8]. Our findings demonstrate novel interplay between local and global structure, charge, and spin states in entropy-stabilized oxides and provide a pathway for tunable magnetic and electronic responses for spintronics and non-filamentary based memristors.

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Molecular Beam Epitaxy Synthesis of LiMn₂O₄ Epitaxial Thin Films

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To address the increasing demand for energy storage technology, model thin films battery cathode systems are highly desirable since they avoid the complexity associated with polycrystalline or nano-sized powders which make detailed study of surfaces and interfaces difficult. However, the behavior of thin films depends highly on defect concentration, grain boundaries, and surface terminations [1,2]. An ideal way to study the material and interface properties is by isolating a particular crystallographic orientation and investigate the orientation dependent performance. To study such behavior, we have developed model thin films systems of different orientations by using molecular beam epitaxy (MBE) to study the interfacial ion diffusion and structural defects and characterized them using aberration corrected transmission electron microscopy (STEM), electron energy loss spectroscopy (EELS), and energy dispersive x-ray spectroscopy (EDXS). These model thin film cathode framework will be used for monovalent Li⁺ ions as well as divalent Mg²⁺ ions intercalation to quantify the evolution of defect concentrations and surface structures.

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Extended defects, substoichiometric phases and defect interactions in oxide based memristive devices

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The emerging “golden age” of artificial intelligence requires energy efficient computing architectures that – in turn – require novel hardware such as memristive systems. A large variety of physical implementations of memristors is currently discussed, including different classes of materials and different working principles. A particular challenge is the transition from digital to analog behavior as required for neuromorphic computing. A major challenge is the correlation of materials properties and device behavior. Defect engineering is the key in semiconductor device physics and technology, and it will also be the key to design novel and adapted memristive systems. One way to realize a memristive device is to induce a reversible soft breakdown along a conducting filament in a dielectric material.

Point defects obviously allow charge carrier doping and substitution to tailor the electric and thermal properties of the dielectric insulator in which the soft breakdown has to be controlled on an atomistic scale. Due to the involved elevated temperatures during operation, even initially amorphous materials will develop a polycrystalline structure. It is well known from materials science, that grain boundaries interact with point defects and may have an important influence on the electronic and thermal properties of the nano-sized functional layer. Furthermore, due to the oxygen dynamics substoichiometric (conducting) phases play an important role.

In this paper we report the effect of oxygen vacancies on the memristive device behavior including compact modelling. One important effect is that oxygen vacancies enhance electric conductivity in the conducting filament and thermal conductivity in the surrounding dielectric matrix. We then show how grain boundaries evolve and how they can be engineered to assume a specific crystal orientation [1]. The grain boundary orientation has a dramatic effect on the forming voltages showing that they have to be taken into account for a full understanding of memristive devices. Furthermore, we investigate the occurrence of substoichiometric phases as a result of the oxygen dynamics [2]. Operando transmission electron microscopy (TEM) plays a key role in atomistic device characterization as needed for model development. Last but not least, we show how defect engineering can be applied to implement analogue depression and potentiation of the resistance in memristive devices, and how the gained knowledge serves as guideline for materials selection [3]. Last but not least, we show that most types of memristors (ferroelectric, phase change and oxide based) are extremely irradiation resistant making them suitable for application in harsh environments and aerospace applications [4].

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Oxide-based nanoelectromechanical sensors and actuators

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Thanks to the rich variety of their physical properties and to their structural and chemical compatibility, complex oxides offer the opportunity to add multiple functionalities to a plethora of devices based on thin film technology. In this presentation I will review some of our recent results in the field of *oxide nanomechanics*, where the peculiar physical properties of selected complex oxides –such as the occurrence of structural or magnetic phase transitions - are exploited through epitaxy for the fabrication of thin film microelectromechanical systems (MEMS). We focus on the fabrication processes of oxide MEMS and on their mechanical characterizations, with particular attention to the role of thin film strain in the device behavior. Some examples of mechanical devices made with (La,Sr)MnO₃ and VO₂ thin films are presented.

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Projects websites: www.oxinems.eu, www.vo2actuators.spin.cnr.it

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Trend in Optical Bandgap of $A_2B_2O_7$ ($A = \text{Sn, Pb}; B = \text{Nb, Ta}$) Thin Films

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Pyrochlore oxides ($A_2B_2O_7$) have been a fruitful playground for condensed matter physics because of the unique geometry in the crystal structure. This crystal structure can be also written as $A_2B_2O(1)_6O(2)$ when two oxygen-sites are explicitly distinguished (Fig. 1(a)). Especially focusing on $A_2O(2)$ tetrahedral sub-lattice in certain pyrochlore oxides $A_2B_2O_7$ ($A = \text{Sn, Pb}; B = \text{Nb, Ta}$), recent theoretical studies predict the emergence of “quasi-flat band” structure because of the strong hybridization between filled A - ns ($\text{Sn-}5s/\text{Pb-}6s$) and O - $2p$ orbitals as shown in Fig. 1(b) [1]. Due to this, this class of compounds is expected to be an ideal platform for realizing numbers of emergent quantum phenomena related with flat band structure, such as ferro-magnetism, high-temperature superconductivity, the fractional quantum Hall effect, and topological states, as in the case of moiré materials. Yet, previous work on bulk samples of the $A_2B_2O_7$ reported that the quasi-flat band was fragile against chemical off-stoichiometry because of the unstable divalent state of the A -site cation.

In this study, we have established the growth conditions of the high-quality $A_2B_2O_7$ films by pulsed laser deposition to elucidate their comprehensive optical properties related with the band structure [2]. Absorption-edge energies, both of direct and indirect band gaps, increase in the order of $\text{Sn}_2\text{Nb}_2\text{O}_7$, $\text{Sn}_2\text{Ta}_2\text{O}_7$, $\text{Pb}_2\text{Nb}_2\text{O}_7$, and $\text{Pb}_2\text{Ta}_2\text{O}_7$. This tendency is successfully explained by considering the energy levels of the constituent elements. Furthermore, a comparison of the difference between the direct and indirect band gaps reveals that $\text{Pb}_2B_2O_7$ tends to have a less dispersive valence band than $\text{Sn}_2B_2O_7$ (Figs. 1(c)-1(e)). These results are consistent with the theoretical predictions and are suggestive of the common existence of the quasi-flat band states in this class of compounds. However, our thin film samples show an insulating behavior, which renders the formidable quantum phenomena undetectable through electrical transport measurements. Together with our previous work on Ti-doping in $\text{Sn}_2\text{Nb}_2\text{O}_7$ [3], our findings will be a guiding principle for band engineering of the “flat-band oxides”.

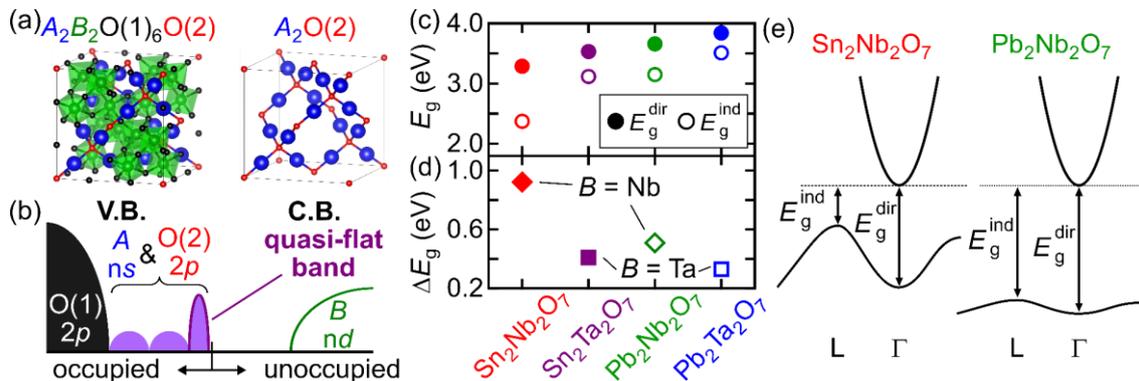


Fig. 1: (a) Pyrochlore, $A_2B_2O_7 = A_2B_2O(1)_6O(2)$, lattice structure and $A_2O(2)$ tetrahedral sub-lattice. (b) Schematics of the band structure. (c) Direct (E_g^{dir}) and indirect (E_g^{ind}) optical bandgaps, and (d) their difference (ΔE_g). (e) Schematics of optical transition for $\text{Sn}_2\text{Nb}_2\text{O}_7$ and $\text{Pb}_2\text{Nb}_2\text{O}_7$ at Γ and L points.

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Study of phonon scattering mechanisms in ferroelectric BaTiO₃ thin films

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The possibility of realizing thermal transport control with ferroelectrics has received increasing interest in recent years [1-4]. This proposed modulation is based on ferroelectric domain walls as electric field-reconfigurable scattering centers for phonons. However, to achieve reliable control, the other scattering mechanisms for phonons in these materials must be carefully studied and decoupled.

Here we report on the thermal conductivity of a series of BaTiO₃ thin films grown by pulsed laser deposition (PLD) on SrTiO₃ 001-oriented substrates. The samples were grown at varying laser fluences, leading to differences in the [Ba]:[Ti] ratio in the samples, enabling to study the effect of scattering produced by point defects (i.e. cationic vacancies).

The ferroelectric state of the samples is corroborated by Raman spectroscopy measurements, showing the presence of the polar BaTiO₃ phase. The structure is probed by means of XRD, showing that samples have relaxed and strained fractions, also visible in TEM images of the samples.

Lastly, the different scattering mechanisms are studied by means of the Debye model [4-7]. The results show the importance of point defect scattering on the thermal transport of BaTiO₃ thin films, as well as the effect of boundary scattering by either domain walls or grain boundaries.

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Understanding the structure-property relationships in relaxor ferroelectric thin films

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Understanding materials structures and how they give rise to their properties is central to materials improvement. In this talk, I will present a series of recent investigations on the structure-property relationships in epitaxial thin films made of the prototypical relaxor ferroelectric $0.68\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\text{-}0.32\text{PbTiO}_3$ (PMN-PT). The presented work is primarily based on the development of epitaxial thin films as a new platform for investigating the behavior of nanoscale-polar structures under different conditions. First, I will present the work on the epitaxial strain dependence of domain structures and dielectric and ferroelectric properties in PMN-PT thin films [1]. We observed that the diffuse-scattering patterns arising from the existence of polar nanodomains change from butterfly shapes to disc shapes as the epitaxial compressive strain increases due to the strain-induced polarization rotation. Even at the highest strains, the films exhibited persistent relaxor behavior due to the enhanced disorder in unit-cells with complex chemistries. Next, I will present the work on the relationship between polarization rotation and polar nanodomains using a multi-scale approach that probes the structural evolution in both the average unit-cells and polar nanodomains [2,3]. We observed that the boundary between (pseudo-)rhombohedral and (pseudo-)tetragonal crystal structures of unit-cells is located in the same region of the strain-electric-field diagram as the boundary between ellipsoidal and cylindrical polar nanodomains, which are elongated along the $\langle 011 \rangle$ and $[001]$, respectively. As the strain increases, the competition between chemistry-driven disorder and strain-driven order of the polar units intensifies, which is manifested in the coexistence of elliptical and cylindrical polar nanodomains in the intermediate step of polarization rotation. These results imply that structural transitions between polar nanodomain configuration underpins the polarization rotation and large electromechanical coupling of relaxors. In addition, I will present the work on the finite size effects in PMN-PT, which led to the construction of a thickness-temperature phase diagram. We observed that the distinguishing relaxor features of PMN-PT disappear when the thickness is below the characteristic size of the polar nanodomains. Ultimately, this implies that the compositional heterogeneities not only determine the size of the PNDs but also play a vital role in defining the stability of relaxors. Lastly, I will demonstrate how the advanced understanding of structure-property relationships afforded by studies presented herein can lead to significant improvements of useful materials properties. Specifically, I explored the enhancement of capacitive energy storage properties in ion-bombarded PMN-PT thin films [5]. This work is based on the observation from the strain dependence that relaxor behavior is surprisingly persistent even in highly ferroelectric environments [1]. The balance between persistent relaxor behavior and reduced electrical leakages in the presence of defects created by ion-bombardment with high-energy helium ions led to unprecedented capacitive energy storage performance.

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Functionality of metastable perovskite ferroelectric CaTiO₃ thin films

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Recently, an exciting new concept based on flexoelectricity has emerged, which uses mechanical forces to switch ferroelectric polarization. This so-called mechanical switching of polarization could be technologically advantageous over conventional switching of polarization via an external electrical bias. First of all, it could minimize electrical bias-induced side effects, such as charge injection, Joule heating, and dielectric breakdown. In addition, it might potentially enable much higher-density data writing, compared to conventional electrical bias-driven polarization switching. In this presentation, we report a breakthrough finding of hyper-efficient mechanical polarization switching in metastable ferroelectrics and overcome the previous dilemma. A combination of density functional theory, phenomenological modeling, and electrical and mechanical characterizations reveals distinctly superior mechanical switching of polarization in an artificial metastable ferroelectric CaTiO₃ film, compared to other conventional ferroelectrics. Furthermore, we demonstrate the phase coexistence and enhance piezoelectricity in a few tens of nanometer thick metastable (111)-oriented ferroelectric thin films via a thickness-driven phase transition.

A multiferroic two-dimensional electron gas

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Multiferroics are compounds in which at least two ferroic orders coexist – typically (anti)ferromagnetism and ferroelectricity [1]– and whose investigation has been a major area of materials science during the last two decades. While magnetic order can arise in both insulating and metallic compounds, ferroelectricity is in principle only allowed in insulators, although ferroelectric metals were proposed over 60 years ago [2]. Recently, several two-dimensional (semi)metals have been reported to behave as ferroelectric [3-5]. Yet, the combination with magnetic order remains elusive. Here we show that a metallic and multiferroic system can be created in an oxide-based two-dimensional electron gas (2DEG) by heterostructure engineering.

A ferromagnetic and a ferroelectric 2DEG has been theoretically modeled and experimentally realized by introducing two-unit cells of the antiferromagnetic insulator EuTiO_3 between LaAlO_3 band insulator and ferroelectric (FE) 1%Ca-doped SrTiO_3 .

By using total electron yield Ti- $L_{3,2}$ edge x-ray linear dichroism we show that the 2DEG orbital splitting and related Ti-O polar displacements are tuned in a non-volatile manner by the FE polarization direction, switched *in-situ*. Eu $M_{4,5}$ edge and Ti- $L_{3,2}$ edge x-ray magnetic circular dichroism show that Eu and Ti- cations order ferromagnetically with an in-plane easy-axis.

Transport data and FE-polarization vs. gate voltage loop show an electric field-induced hysteresis of the sheet resistance correlated to the FE-polarization and an anomaly of the R_{sheet} vs T dependence at the FE T_c . Furthermore, the 2DEG displays an anomalous Hall effect and a magnetoresistance that can both be modulated and cycled by switching the remanent polarization, demonstrating a magnetoelectric coupling.

Our findings provide new opportunities in quantum matter stemming from the interplay between ferroelectricity, ferromagnetism, and Rashba spin-orbit coupling in an oxide 2DEG.

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Field-induced reversal of paramagnetic Nd moments in ferromagnetic $\text{Nd}_2\text{NiMnO}_6/\text{La}_2\text{NiMnO}_6$ superlattices

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We investigate the magnetic behavior of superlattices of the double perovskites $\text{Nd}_2\text{NiMnO}_6$ and $\text{La}_2\text{NiMnO}_6$. Both compounds feature ferromagnetism induced by super exchange between the B-site-ordered Ni and Mn ions with a T_C of 200 K and 280 K, respectively [1, 2]. Using a custom-built RF magnetron sputtering setup equipped with in situ RHEED we grow superlattices of different periodicities and fixed total thickness, Figure 1 (a). We grow x unit cells $\text{Nd}_2\text{NiMnO}_6$ and $\text{La}_2\text{NiMnO}_6$ each which is repeated y times; denoted as (x,y) . SQUID $M(T)$ measurements show two distinct para to ferromagnetic transitions for high periodicity superlattices. Intriguingly, for samples with a low periodicity the two transitions combine and feature a single T_C right between those of the individual compounds. Further, at low temperature, we observe two additional openings in the ferromagnetic loops of the low periodicity superlattices in SQUID $M(H)$ measurements, Figure 1 (b). To understand this striking behavior, we resort to element-specific XMCD measurements for an exemplary $(3,3)_{10}$ superlattice, Figure 1 (c). The Ni and Mn moments manifest the expected ferromagnetic behavior. Strikingly, however, the Nd moments feature a non-saturating paramagnetic behavior and a reversal at low applied fields compared to the Ni/Mn moments, causing the intriguing shape of the SQUID $M(H)$ loops. The fact that this is not observed in the high periodicity samples and noting that all samples contain the same total amount of Nd highly affirms that this effect is interface driven.

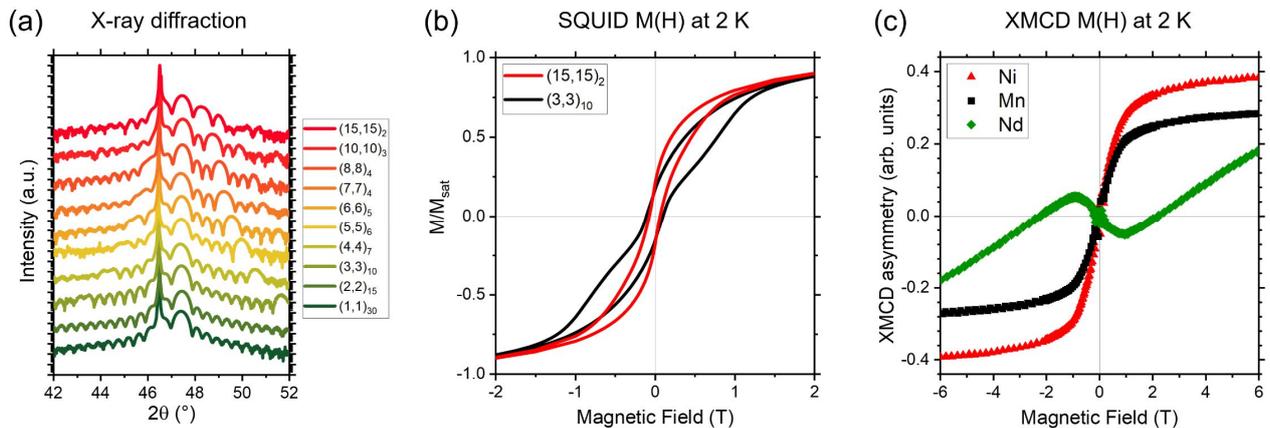


Figure 1 (a) X-ray diffraction for superlattices of different periodicities around the SrTiO_3 (002) substrate peak. Laue oscillations indicate high crystal quality, and additional superlattice satellite peaks demonstrate the exceptional interface quality between the two constituents. (b) SQUID $M(H)$ measurements for two superlattices of different periodicities. The low periodicity $(3,3)_{10}$ sample features two additional openings not present in the high periodicity sample. (c) XMCD $M(H)$ measurements on the $(3,3)_{10}$ superlattice at the Ni, Mn, and Nd absorption edges. The Nd moments show a paramagnetic behavior with a sign reversal at low applied fields.

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Controlling Ferroelectricity and Quadruple-well State in CuInP_2S_6 via Interfacial Epitaxial $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$

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Layered van der Waals CuInP_2S_6 (CIPS) exhibits room-temperature ferroelectricity with unconventional quadruple-well states. The high mobility of Cu ions in this two-dimensional (2D) semiconductor makes it challenging to achieve nanoscale control of its polarization. Here we report the enhanced domain formation and piezoelectric response of CIPS via the interfacial charge coupling with an epitaxial ferroelectric oxide. We transferred mechanically exfoliated 8-300 nm CIPS flakes on top of Si, Au, and 50 nm epitaxial (001) $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT) films. Piezoresponse force microscopy (PFM) studies revealed random domains formed in CIPS flakes on Si and Au substrates, while thin flakes (< 25 nm) on PZT exhibit uniform polarization. Figures 1(b)-(c) show that the polar alignment effect in CIPS vanishes with increasing CIPS thickness (t_{CIPS}). To quantify the thickness dependence of the coupling effect, we extracted the piezoelectric coefficient d_{33} of the CIPS flakes (d_{33}^{CIPS}) on the three types of substrates. The change of d_{33}^{CIPS} on PZT can be divided into three regions: the negative d_{33} region I ($t_{\text{CIPS}} > 50$ nm), the intermediate region II ($25 < t_{\text{CIPS}} < 50$ nm), and the positive d_{33} region III ($t_{\text{CIPS}} < 25$ nm). Compared with the results on Si and Au, the magnitude of d_{33} for thin CIPS on PZT (region I) is significantly enhanced, and the sign of d_{33} changes to positive. We also performed *in-situ* PFM studies at elevated temperatures and observed a Curie temperature of ~ 200 °C for CIPS on PZT, which is much higher than the previously reported value for bulk CIPS (87 °C) [1].

The unconventional evolution of d_{33}^{CIPS} can be attributed to the position shift of Cu cation between the metastable and ground state in the quadruple-energy-well, which is driven by the interfacial charge coupling with PZT. Density functional theory calculations revealed that CIPS and PZT have their polarizations antialigned due to the accumulated charges at the interface. Our study points to a new strategy to engineer the nano-domain structure and piezoelectric response of the 2D ferroelectric CIPS.

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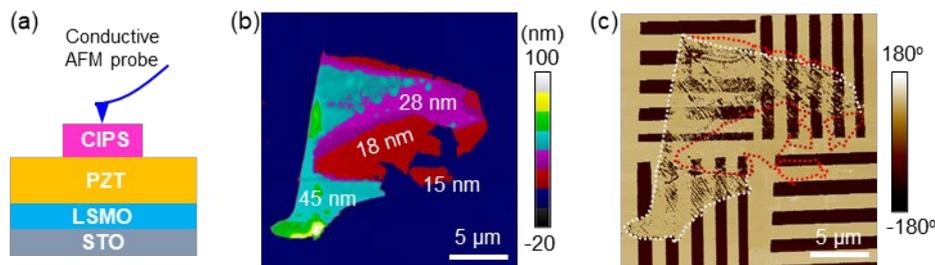


Figure 1 (a) Schematic experimental setup. (b) Topography and (c) PFM phase images of CIPS flakes on the pre-patterned domains of PZT.

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Can BaFe₁₂O₁₉ be Transmuted into a Room-Temperature Ferrimagnetic Ferroelectric?

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While electric-field control of magnetism is a promising way to achieve ultralow-power logic-memory devices, it poses a grand challenge for science. Ideal materials for this application need to have strongly coupled electric and magnetic orders at room temperature, high spontaneous magnetization, and low leakage [1]. Unfortunately, no known single-phase multiferroic meets all of these criteria. For example, the Achilles' heel of BiFeO₃, the most widely studied multiferroic material, is its low spontaneous magnetization (about 0.03 μ_B per unit cell).

To construct a single-phase multiferroic material with better magnetic properties, we are trying to transmute a ferrimagnetic, incipient ferroelectric material, BaFe₁₂O₁₉, into a room-temperature ferrimagnetic, ferroelectric via epitaxial strain. BaFe₁₂O₁₉ possesses a net magnetic moment of 40 μ_B per unit cell and has a high magnetic Curie temperature of 723 K [2]. Long recognized as an incipient ferroelectric [2], BaFe₁₂O₁₉ is predicted to transform into a ferroelectric state at room temperature when more than 4% in-plane biaxial compressive strain is applied [3]. Here we show that a 1.1% in-plane biaxial compressive strain from a newly developed substrate [4] can be imposed on a 15 nm thick BaFe₁₂O₁₉ film. The full width at half maximum (FWHM) of the X-ray diffraction rocking curve of the 0014 peak in ω is 0.009°, the smallest ever reported. Our preliminary scanning transmission electron microscopy (STEM) results on a commensurately strained film show local electric polarization arising from the off-center displacement of Fe³⁺ ions in the trigonal bipyramid sites, locally breaking the mirror-plane symmetry perpendicular to the *c*-axis of BaFe₁₂O₁₉.

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Bond-dependent slave-particle cluster theory based on density matrix expansion

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June 15, 2022

Solving large and strongly-correlated electronic problems, exemplified by the Hubbard model on a lattice, is an ongoing electronic structure challenge, and a wide variety of approaches exist in the field. Recent occupation-number based slave-particle methods [1-4] represent a computationally efficient approach for obtaining approximation solutions. In these methods, one decouples the electronic spin and charge degrees of freedom to end up with an easy non-interacting fermion problem on the lattice and an easier interacting auxiliary slave problem on the lattice. However, one must then truncate or simplify the slave problem to render it tractable computationally.

We introduce a new cluster slave-particle method for Hubbard models describing transition metal oxides. The method is based on the expansion of the lattice density matrix into a set of overlapping clusters in real space. Our approach includes all the nearest-neighbor hopping terms directly within the interacting clusters and does not truncate or approximate them at cluster boundaries (unlike prior cluster approaches including cluster DMFT [5-7]). The approach also overcomes some of the shortcomings of prior single-site slave-particle methods (e.g., predicting a false Mott transition in one dimension). We test our approach on 1D and 2D $p-d$ model systems and compare to numerically exact results based on exact diagonalization and density matrix renormalization group (DMRG). We find that our approach is computationally economical and high-quality total energies, site occupancies and d -site double occupancies as a function of Coulomb interaction strength and doping.

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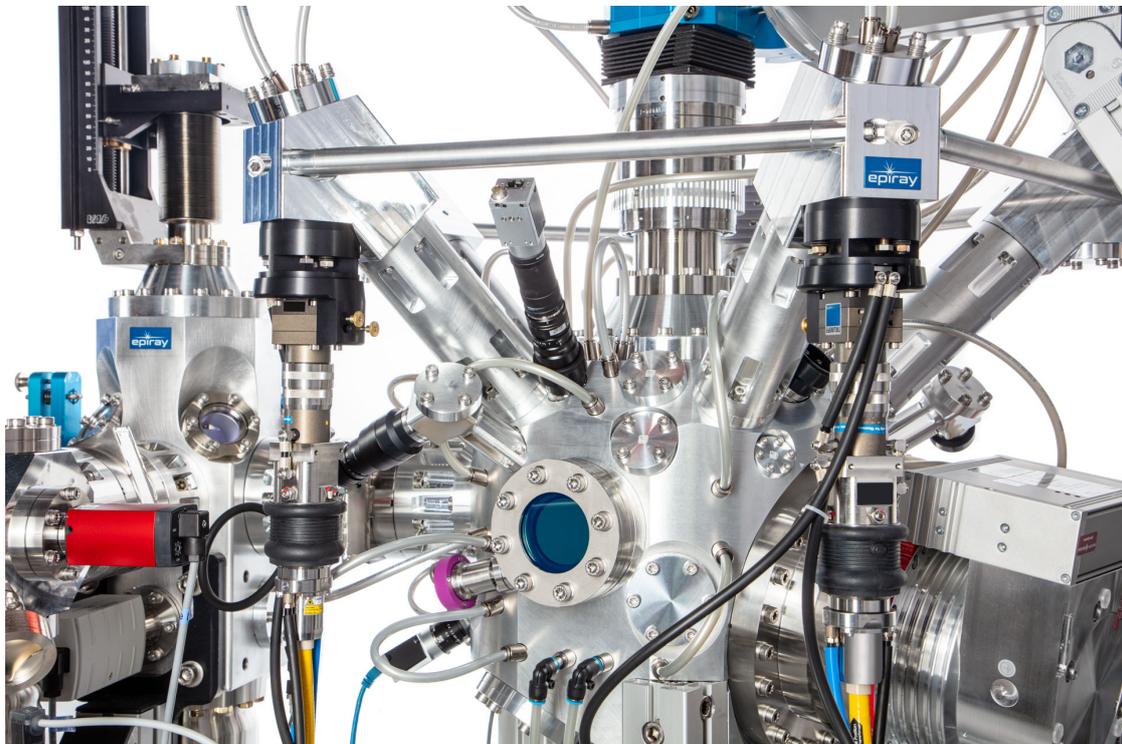
Next Generation Epitaxy: clean, simple, fast and versatile

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¹ Epiray GmbH, Heisenbergstrasse 1, Stuttgart, Germany

Epiray introduces **Thermal Laser Epitaxy (TLE)** – a novel technology for the deposition of high-purity thin films. Epiray is a spin-off of the Max Planck Society pioneering a new era of epitaxy research.

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Photograph of a TLE system.

TLE expands both the temperature and oxygen pressure ranges at which source material can be evaporated. Furthermore, the TLE system is equipped with a laser substrate heater that can operate up to temperatures above 2000 degrees Celcius. This dramatically increases the available parameter space for epitaxy. TLE is thus the ideal method for creating **high-quality oxide heterostructures**.

Investigation of carbon monoxide adsorption on In_2O_3 surface for gas sensing applications

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In_2O_3 is n-type wide band-gap semiconducting oxide, and is promisingly used as gas sensing device. The principle of gas detection of such devices is based on change in electrical conductance upon exposure to the gases. An attempt has been made to understand the phenomena taking place on In_2O_3 surface due to adsorption of gases. A melt-grown bulk In_2O_3 single crystal with an as-cleaved (111) surface was used for the present study. Scanning electron microscopy and spectroscopy (STM/S) measurements exhibit the changes in structural and electronic properties due to adsorption of varying doses of carbon monoxide.

Alcohol Sensing Properties of CuO-CNT Hybrids

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The present work reports the effect of introducing carbon nanotubes into copper oxide on structural, morphological, optical and alcohol sensing properties. To have insight into the structural, morphological and optical properties of the synthesized samples, various characterization techniques such as X-ray diffraction, transmission electron microscopy and UV-Visible spectroscopy were employed. The sensors were fabricated out of the synthesized samples to test their response towards methanol, ethanol and propanol. The synthesized CuO-CNT sensors exhibit enhanced sensing performance, and are capable of detecting concentration of alcohol as low as 10 ppm at 300 °C.

Cubic epitaxial perovskite SrTaO₃ for optoelectronic applications

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Abstract

The ever wide increase in the demand of consumer electronics is playing a large role in the development and realization of a new class of materials whose properties are rarely observed in the nature. While theoretical methods like density functional theory (DFT) combined with machine learning [1] have predicted a whole new set of compounds and materials which might show promising results in the future, the experimental findings and fabrication are still playing catch up. In this regards, the set of materials whose properties have been promising but not yet fully studied and understood, is growing larger day by day. One such problem is finding a viable replacement for the Tin doped Indium Oxide (ITO). While a large number of compounds have been predicted, they fall short due to a large number of factors. One of them, SrNbO₃ [2] [3] was shown to be promising, but its optical properties are less than desirable. A new isoelectronic compound to the SrNbO₃, SrTaO₃ has been proposed [4]. SrTaO₃ is a 5d transition metal oxide (TMO) with strong correlations. While previous theoretical modelling has predicted anticipated properties, the epitaxial growth has been challenging so far. 5d TMOs such as SrIrO₃ [5] and SrVO₃ [6] have already been studied extensively and shown exceptional range of properties, from ferromagnetism to two dimensional electron gas (2DEG) to ferroelectricity etc. In this report, SrTaO₃ thin films are grown on cubic substrate, LaAlO₃ epitaxially using pulsed laser deposition (PLD) and their optoelectronic studies are undertaken. DFT studies provide us with the extra details that are needed to understand the system in a complete way. The subsequent studies of the films are done using high-resolution X-ray diffraction (HR-XRD) for the structural properties. Then the surfaces are characterised via atomic force microscopy (AFM). Optical studies are done using spectroscopic ellipsometry and UV-Vis spectroscopy. The electrical and magnetic studies are done using physical property measurement system (PPMS). The confirmation of epitaxially grown films are first demonstrated and other studies determine its potential for use as an optoelectronic device.

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Low resistance epitaxial edge contacts to buried nanometer thick conductive layer of La-doped BaSnO₃

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As the size of the semiconductor device decreases, the importance of the low resistance contacts to devices cannot be overstated. Here we studied the contact resistance to a buried nanometer thick δ -doped Ba_{1-x}La_xSnO₃ (BLSO) layer [1]. We have used epitaxial 4% (x=0.04) BLSO as a contact material which has additional advantages of forming ohmic contacts to BaSnO₃ and providing thermal stability even at high temperatures. The contact resistance was measured by a modified transmission line method designed to eliminate the contribution from the resistance of the contact material. The contact resistance to a 5.2 nm thick δ -doped 1% BLSO conductive layer was measured to be smaller than $4.13 \times 10^{-6} \Omega\text{cm}^2$. Our results show that it is possible to provide low resistance epitaxial contacts to an embedded nanometer-thick BLSO conductive layer using an ion-milling process. Our low resistance contact method can be easily extended to a 2-dimensional electron gas (2DEG) at the oxide interfaces such as LaInO₃/BaSnO₃

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Stress Analysis and Q-Factor of Free-Standing (La,Sr)MnO₃ Oxide Resonators

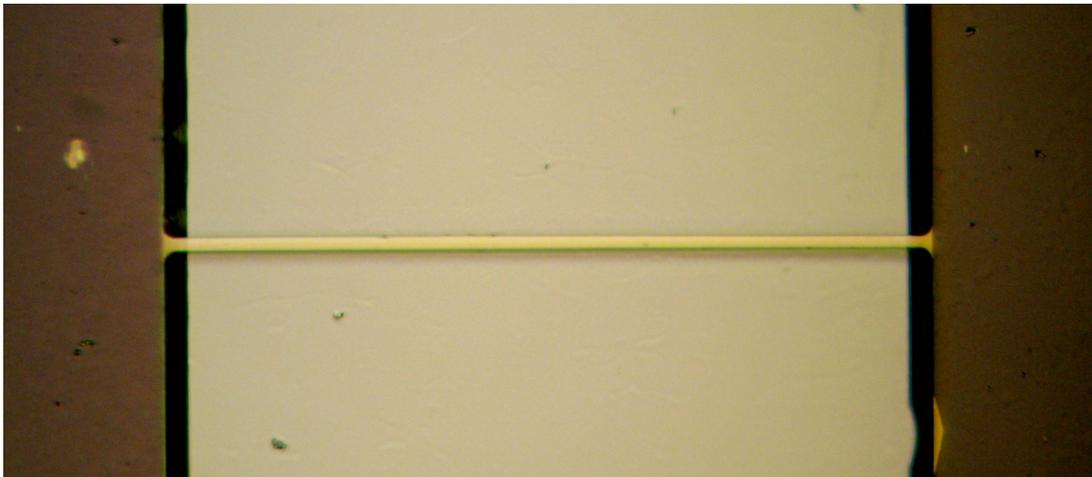
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High-sensitivity nanomechanical sensors are mostly based on silicon technology and related materials. The use of functional materials, such as complex oxides having strong interplay between structural, electronic, and magnetic properties, may open possibilities for developing new mechanical transduction schemes and for further enhancement of the device performances. Nevertheless, the integration of these materials into micro-mechanical transducers is still at its very beginning and critical basic aspects related to the stress state and the quality factors of micro/nanoresonators made from epitaxial oxide thin films need to be investigated. We will discuss the fabrication issues and the mechanical characteristics of suspended micro-bridges made of single-crystal thin films of (La_{0.7},Sr_{0.3})MnO₃ (LSMO), a prototypical complex oxide showing ferromagnetic ground state at room temperature. These mechanical resonators result to be highly stressed, with strong temperature dependence of the resonance frequency around the magnetic transition. Their Q-factors reach few tens of thousands at room temperature, with indications for a further improvement by optimizing the fabrication protocols [1]. Our results demonstrate that complex oxides can be employed to realize high Q-factor mechanical resonators, paving the way towards the development of full-oxide MEMS/NEMS sensors.



A 200 μ m-long suspended bridge fabricated from a 100nm-thick film of single-crystal LSMO deposited on top of SrTiO₃(110)

References (Times New Roman 10 pt; bold)

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Mechanical properties of epitaxial EuTiO_3 thin-film MEMS resonators

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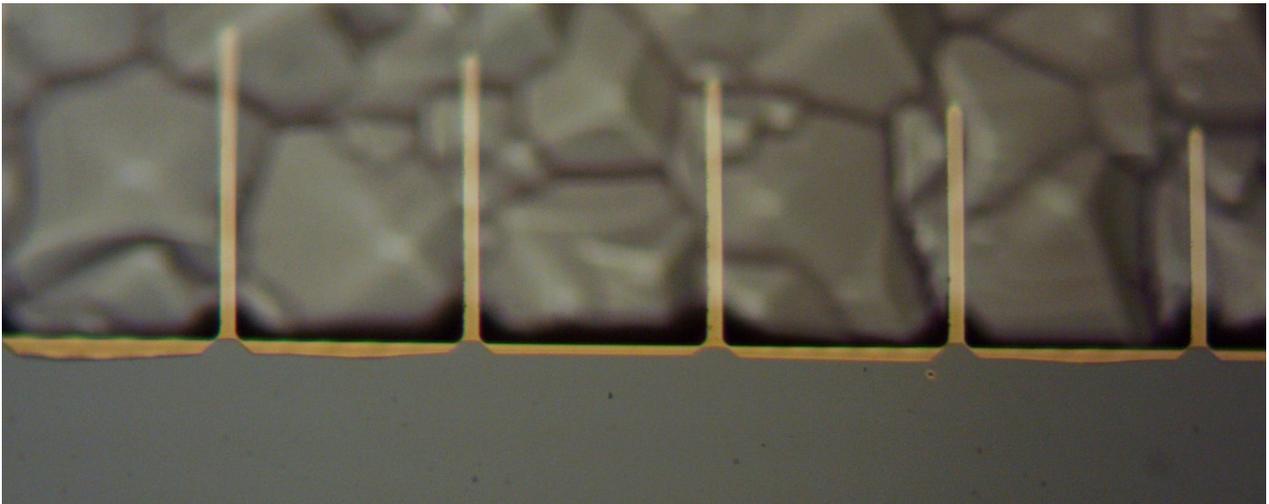
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The fabrication of suspended structures based on complex oxides enable the realization of new experiments and devices where mechanical deformations, thermal insulation, and interaction with external fields. In this framework, designing a material library for developing different types of structures with oxides is mandatory to enrich the design capabilities of micro and nano-mechanical devices. EuTiO_3 is a transparent dielectric with lattice parameters proximal to SrTiO_3 . We will show the mechanical characterization of EuTiO_3 suspended cantilever and micro-bridges, that allowed us to evaluate the strain state of the epitaxial thin film as well as its Young's modulus. These results provide the basic characterizations required to consider EuTiO_3 as a new brick for the realization of more complex full-oxide MEMS devices.



EuTiO_3 cantilevers (5 μm -wide) fabricated from epitaxial thin film deposited on top of $\text{SrTiO}_3(100)$

Voltage control of spatially-patterned functional responses in lateral perovskite heterostructures via ionic gating

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The drive to control functional properties using electric fields has been a motivating theme within the oxide heterostructure community for the last two decades. To this end, electrolytic gating is a proven approach for modifying properties such as magnetism or superconductivity, including through the insertion and removal of oxygen anions in films [1]. Here, we show how ionic gating applied to lateral oxide/oxyfluoride heterostructures enables reversible voltage control over a transition between metal/insulator and insulator/insulator lateral patterns, leading to electric field modulation of anisotropic transport and optical responses. Lateral stripes of SrFeO_{2.5}/SrFeO₂F are patterned into ferrite films on conducting SrTiO₃:Nb substrates through lithographically-defined topochemical fluorination reactions [2]. Using a back-contact geometry, biasing across an ionic gel results in a conversion of the SrFeO_{2.5} regions to SrFeO₃, while SrFeO₂F remains unchanged after biasing. Thus, the application of an oxidizing bias ($V_G = -3$ V) converts the all-insulating SrFeO_{2.5}/SrFeO₂F pattern into SrFeO₃/SrFeO₂F metal/insulator lateral structures with highly anisotropic in-plane transport and strong optical contrast between stripes. The application of a positive bias ($V_G = 1$ V) results in reduction of the SrFeO₃ phase. As opposed to simple reduction to SrFeO_{2.5}, we provide x-ray diffraction and photoemission results indicating that the positive bias leads to the formation of a vertical SrFeO_{2.5}/HSrFeO_{2.5} bilayer with the protonated phase buried at the film/substrate interface (the SrFeO₂F regions exhibit no change after this positive bias). These bilayer regions can be reversibly converted back to the SrFeO₃ perovskite phase with a negative gate bias. This work thus expands the scope of properties that can be controlled via electrolytic gating to include macroscopic anisotropic transport behavior and optical responses that rely on in-plane dielectric contrast [3]. We anticipate that this approach for realizing voltage-control over patterned properties will be broadly applicable to oxide materials amenable to fluorination that host oxygen-vacancy-dependent properties.

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All-oxide thin-film varactors with Mn- and Ni-doped (Ba,Sr)TiO₃ for microwave applications

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Tunable microelectronic devices operating over a wide range of frequencies are required for mobile communications, high-speed data connections with the upcoming 5G and Internet of Things (IoT) technologies. A key requirement of such devices is the reduction of dielectric losses. Oxide electronics offers novel routes to avoid losses in the substrate and in the dielectric layer by using epitaxial stacks on extremely thick ultra-highly conducting SrMoO₃ based bottom electrodes that can be grown coherently strained with several μm thickness [1]. Here, we focus on the improvement of the fully epitaxial dielectric layer. The possibility to use highly crystalline dielectric layers suppresses defect interactions and extended defects and allows studying the role of dopants in the bulk of the dielectric material without the interference of e.g. grain boundaries.

Here we present all-oxide thin-film tunable capacitors (varactors) with single-crystalline films of a tunable dielectric perovskite Ba_xSr_{1-x}TiO₃ (BST) doped with Mn and Ni, grown epitaxially by pulsed laser deposition on SrMoO₃ bottom electrodes with a room-temperature resistivity of 30 $\mu\Omega\text{cm}$. Partial substitution of the tetravalent Ti at B-sites of the BST perovskite structure with Mn and Ni in a trivalent or divalent state decreases the concentration of charge carriers due to oxygen vacancies, leading to a reduction of the leakage current density of the varactor by 5 orders of magnitude down to 1 A/m². The low leakage current allows decreasing the BST thickness in all-oxide varactors below 50 nm, enabling ultra-low tuning voltages – a unique feature of this particular oxide device.

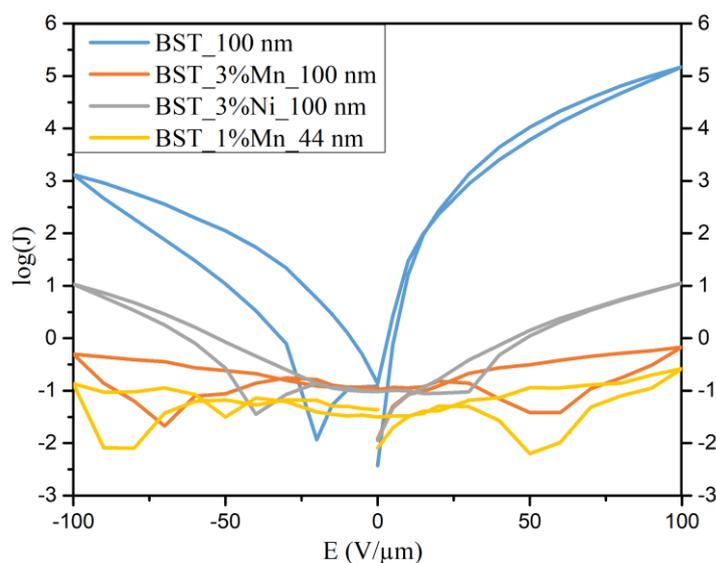


Figure 1. Leakage current of the Pt/BST/SrMoO₃ varactors with tunable BST, Mn- and Ni-doped BST functional layers.

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CoFeB/Y₃Fe₅O₁₂ bilayer magnonic resonator for magnetic sensor application

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Spin waves (SWs)—which is a collective excitation of electron spin, can be manipulated by a wide range of parameters; thus, they are suitable for sensing applications in a wide range of physical fields [1]. In this study, we designed a highly sensitive, simple structure, and ultra-low power magnetic sensor using a simple CoFeB/Y₃Fe₅O₁₂(YIG) bilayer structure. We grew a 70-nm-thick YIG thin film on a Gd₃Ga₅O₁₂ substrate using the PLD technique. 60 nm thick and 15 μm wide CoFeB stripe was patterned onto the YIG films by photolithography and room-temperature DC magnetron sputtering. Subsequently, a system of two CPWs composed of 90-nm-thick Au was integrated into the YIG film using photolithography and DC magnetron sputtering. We employed a microwave characterization technique using a vector network analyzer connected to two coplanar waveguides labeled CPW1 and CPW2 shown in Figure 1(a) and (b). A DC-biased magnetic field ($\mu_0 H_{\text{ext}} = 20$ mT) was applied where magnetostatic surface SWs were excited. We demonstrated that the CoFeB/YIG bilayer structure can create a sharp rejection band in its spin-wave transmission spectra shown in Figure 1(c) similar as [2]. The lowest point of this strong rejection band allows the detection of a small frequency shift owing to the external magnetic field variation demonstrated in Figure 1(d) and (e). Experimental observations revealed that such a bilayer magnetic sensor exhibits 20 MHz frequency shifts upon the application of an external magnetic field of 0.5 mT. Considering the full width half maximum of the rejection band, which is about 2 MHz, a sensitivity of 10⁻² mT order can be experimentally achieved. Furthermore, the higher sensitivity in the order of 10⁻⁶ T (μT) has been demonstrated using the sharp edge of the rejection band of the CoFeB/YIG bilayer device shown in Figure 1(f). We proposed an Y-shaped SWs interference device with two input arms consisting of CoFeB/YIG and YIG to demonstrate a sensitivity of 10⁻⁹ T (nT) at room temperature shown in Figure 1(g).

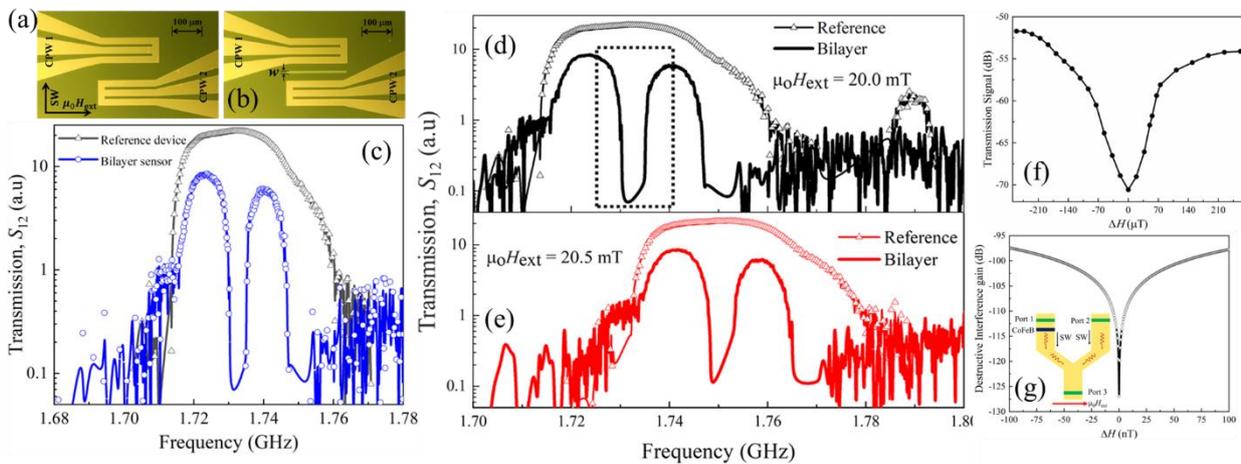


Figure 1. Optical image of (a) reference and (b) CoFeB/Y₃Fe₅O₁₂ bilayer resonator (c) Transmission spectra of reference and bilayer resonator. Transmission spectra of reference and bilayer resonator at (d) 20 mT and (e) 20.5 mT bias magnetic field (f) Evidence of micro-Tesla sensitivity using bilayer resonator. (g) nano-Tesla sensitivity prediction using interference and bilayer resonator combination.

This work was partially supported by JSPS KAKENHI Grant Number 20H05651, 19K15022, 21J12725 and Basic Research Grant (Hybrid AI) of Institute for AI and Beyond for the University of Tokyo.

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High-k perovskite gate oxide for modulation beyond 10^{14} cm^{-2}

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Relentless scaling down of semiconductor devices requires a new class of high-k dielectric materials to continue lowering the operating voltage of field effect transistors (FETs) and storing sufficient charge on a smaller area. Here we have investigated the dielectric properties of the epitaxial $\text{BaHf}_{0.6}\text{Ti}_{0.4}\text{O}_3$ (BHTO), an alloy of perovskite oxide BaHfO_3 and BaTiO_3 . We found the dielectric constant, the breakdown field, and the leakage current to be 150, 5.0 MV cm^{-1} , and $10^{-4} \text{ A cm}^{-2}$ at 2 MV cm^{-1} , respectively. The results suggest that 2-dimensional carrier density of more than $n_{2D} = 10^{14} \text{ cm}^{-2}$ could be modulated by the $\text{BaHf}_{0.6}\text{Ti}_{0.4}\text{O}_3$ gate oxide. We demonstrate an n-type accumulation mode FET transistor and direct suppression of more than $n_{2D} = 10^{14} \text{ cm}^{-2}$ via an n-type depletion mode FET. We attribute the unusual property of large dielectric constant, high breakdown field, and low leakage current of $\text{BaHf}_{0.6}\text{Ti}_{0.4}\text{O}_3$ to the nanometer scale stoichiometric modulation of Hf and Ti.^[1]

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High-quality spinel γ -Fe₂O₃ epitaxial thin film grown on Nb:SrTiO₃ substrate for spin-wave modulation and computation

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Achieving ultra-low energy spintronics applications requires a material that can efficiently propagate spin waves (SW). Spinel-type ferrite is a promising material from the point of view of application due to its potential of great semiconducting performance, it is high promising to realize modulation of spin wave by applying bias voltage or laser. In this study, we fabricated high-quality epitaxial spinel ferrite on conductive Nb:SrTiO₃ substrate and realized spin-wave propagation on it.

We grew a 30-nm-thick Fe₃O₄ thin film on a Nb:SrTiO₃ substrate using the PLD technique. To maintain high-quality cubic spinel structure with a FWHM of 0.1 in rocking curve measurement, samples were annealed at 300 °C for 5 hours in the air. The XRD figure is shown in Fig. 1 (a), we can find there is no second phase existing, which indicates the purity of the thin films. We can conclude that after annealing, the peaks of samples shifted to the right, and it is consistent with the reported results that the lattice constant of Fe₃O₄ and γ -Fe₂O₃. The XPS result is shown in the inset of Fig. 1 (a), this also signals that after appropriate annealing, the Fe³⁺ ions dominate absolutely, which indicates the Fe₃O₄ sample is oxidized fully to turn to Fe₂O₃.

We employed a microwave characterization technique using a vector network analyzer (VNA) connected to two coplanar waveguides shown in the inset of Fig. 1 (b). A DC-biased magnetic field was applied where magnetostatic surface SWs (MSSWs) were excited. We demonstrated that the γ -Fe₂O₃ thin film can create a sharp rejection band in its spin-wave transmission spectra shown in Figure 1(b). It is revealed that the lowest point of this strong rejection band shifts based on the external magnetic field, while the frequency increases with the increase in the magnetic field.

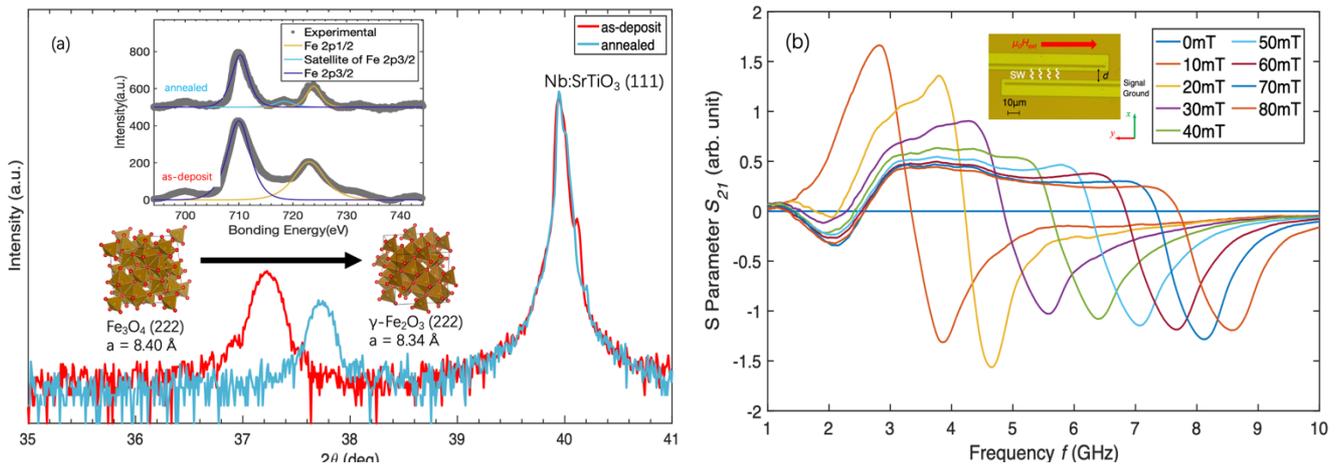


Figure 1 Structure, component, and SW propagation situation of 30 nm γ -Fe₂O₃ thin films. (a) Out-of-plane XRD and XPS measurement of as-deposited and annealed sample. (b) SW propagation measured by vector network analyzer.

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Atomic-scale insights into the structural transformations in cathodes for multivalent metal-ion batteries

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Multivalent-metal ion batteries are promising and safer alternatives to lithium-ion batteries for next-generation energy storage devices. Most of the multivalent-metal ions, such as calcium (Ca) and magnesium (Mg), and their respective cathode material systems can be sourced from earth-abundant minerals. Moreover, Ca and Mg-ion batteries, based on high-voltage oxide cathodes, show high energy density compared to lithium-ion batteries. This makes multivalent-metal ion batteries a cost-effective proposition for a variety of battery technologies. However, multivalent-metal ion batteries often suffer from poor electrochemical stability. A key challenge in achieving higher performance and stability in these material systems is to understand the fundamental structural changes to the cathode at the atomic scale. Such structural changes during electrochemical cycling include cation disorder, planar defects, phase separation, and (de)intercalation mechanisms. In this work, we combine aberration-corrected scanning transmission electron microscopy (STEM), first-principles density functional theory (DFT) calculations, and STEM simulations to develop a fundamental understanding of the structural changes occurring during electrochemical cycling of Ca and Mg-ions into FePO₄ and α -V₂O₅ as cathodes respectively.

During electrochemical cycling of Ca into FePO₄, we observe that the large particles (> 100 nm) show Ca intercalation at the surface with the bulk of the particle showing little to no activity. Moreover, the smaller particles (< 50 nm) exhibit a tendency to phase-separate into CaPO₄. STEM imaging reveals the direct atomic-scale evidence of Ca intercalation into the FePO₄ olivine structure, where Ca-ions show a preferential arrangement along the [010] orientation. For the Mg-ion batteries, we observe that the Mg intercalation into α -V₂O₅ is heterogeneous and is accompanied by the formation of a network of low-angle grain boundaries. We hypothesize that there is significant phase separation between various phases of Mg_xV₂O₅ and V₂O₅, such as the fully intercalated δ -MgV₂O₅ and fully deintercalated α -V₂O₅ phases. Our atomic-scale findings for these two model systems can be leveraged by optimizing the materials synthesis to achieve theoretical operating potentials and performance.

Acknowledgments: This work is supported by the Joint Center for Energy Storage Research (JCESR) and Energy Innovation Hub funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences.

From Schottky to ohmic (La,Sr)MnO₃ / (Ba,Sr)TiO₃ contact by interface engineering

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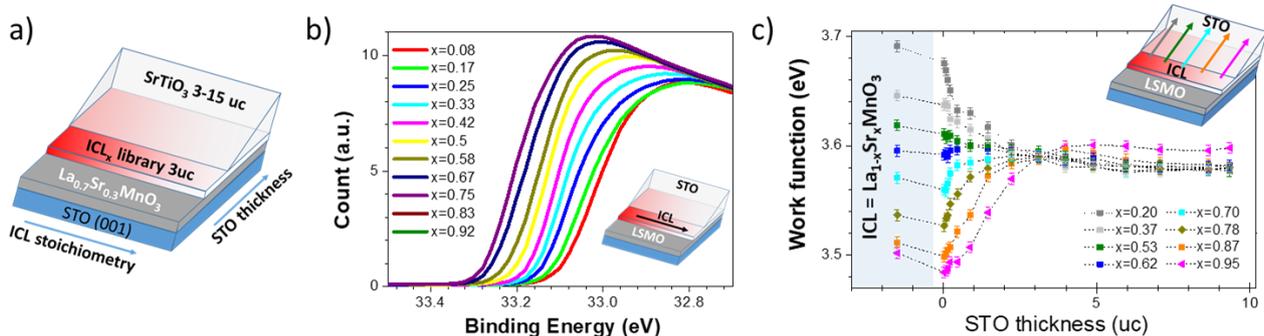
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Band structure engineering in silicon-based heterostructures dedicated to microelectronic and energy harvesting applications has been common material scientist playground for decades. Careful design of the energy level variation at hetero-interfaces is used e.g. to collect electron while preventing electron-hole recombination in PV heterostructures, control electron injection with Schottky barriers (SB), promote quantum well... Although band structure engineering in Si or GaAs based heterostructures is a very mature field, it is a relatively new territory for oxide electronic where emerging new device concepts based on perovskite-derived heterostructures makes it very desirable. In all perovskite-based heterostructures, the structural continuity at ABO₃ hetero-interfaces gives extra degrees of freedom to tune electronic and structural properties. Rumpling, polar discontinuity, interfacial B-site cation environment asymmetry, BO₆ octahedral rotations are all potential levers to alter the band structure and promote electronic properties like e.g. interface polarization¹ or enhanced SB height^{2,3}.

One way to play with these degrees of freedom is to modulate the interface chemical composition by introducing an Interface Control Layer (ICL), as it has been proposed decades ago for Si and GaAs based heterostructures.³ To accelerate the exploration of ICL perovskite materials, we implemented an interface combinatorial pulsed laser deposition (ICPLD) set-up.⁴ We report here on epitaxial La_{0.7}Sr_{0.3}MnO₃/ ICL (3uc) /SrTiO₃ junctions with ICL = La_{1-x}Sr_xMnO₃ and Ba_{1-y}Sr_yTiO₃ (fig. a). XPS/UPS spectroscopies were used to probe core levels, work function (W_F)



and band bending versus ICL composition and STO thickness. Modulation of W_F (fig. b) leading to a transition from Schottky to ohmic contact with the polar discontinuity (fig. c) will be discussed together with interface atomic and chemical structure characterized by HR-STEM technics.

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Exploring free-standing $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ membrane and its nonvolatile gating effect in two-dimensional MoS_2

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Free-standing ferroelectric oxide membranes have attracted considerable research interests recently for its potential to be integrated with other functional materials with distinct crystal symmetry, such as two dimensional (2D) van der Waals materials. It also presents a different disorder energy landscape compared with the epitaxial films, which can significantly affect the static configuration and dynamic response of the ferroelectric domain wall (DW) [1]. Here, we report a comprehensive study of single crystalline free-standing $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT) and PZT/(La,Sr)MnO₃ (LSMO) membranes on different base-layers, including Au, LSMO/STO, and 2D semiconductor MoS_2 . We deposit 10 to 50 nm epitaxial PZT thin films on $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) or LSMO/SAO buffered SrTiO_3 (STO) substrates using off-axis RF magnetron sputtering. The as-deposited heterostructures are single crystalline with no impurity phases, as confirmed by x-ray diffraction measurements. By dissolving the SAO buffer layer in water, we obtain suspended oxide membranes, and transfer the samples on Au and LSMO/STO (Figs. 1a-b). We create stripe domains and (Fig. 1c inset) and analyze the DW roughness via piezo-response force microscopy (PFM) studies. The extracted DW roughness exponent suggests that the PZT and PZT/LSMO membranes transferred on LSMO/STO substrates are dominated by the 2D random bond (RB) disorder, which is similar to that observed in epitaxial PZT thin films [2,3]. In contrast, the membranes transferred on Au substrates show 1D RB disorder dominated behavior (Fig. 1c), which may be attributed to the enhanced rippling effect [4].

We then transfer 50 nm PZT membranes on 2D MoS_2 field effect transistors as the top gate. The ferroelectric polarization either accumulates or depletes the charge carriers in the 2D channel. Switching the polarization induces nonvolatile switching of the channel current, with a high current on/off ratio of $\sim 2.04 \times 10^5$ achieved at room temperature. Both current on/off states show robust retention for over 3 days, making it promising for developing flexible nonvolatile memory applications.

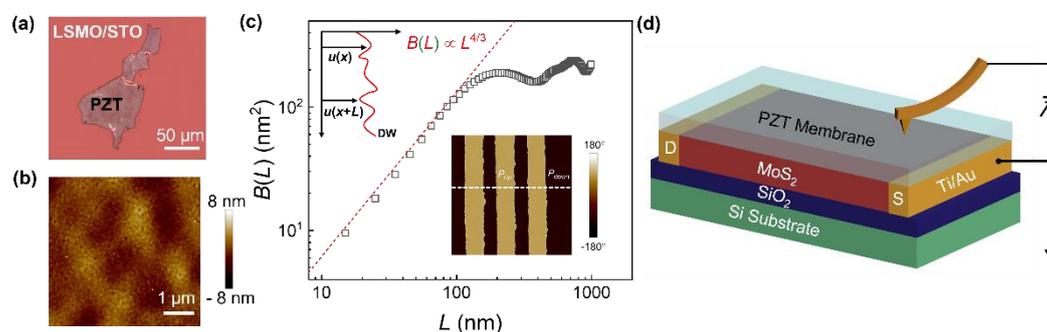


Figure 1. (a) Optical image and (b) AFM topography of 50 nm PZT membrane on LSMO/STO. (c) DW correlation function $B(L)$ of 50 nm PZT membrane on Au/ SiO_2 /Si substrate. The dashed line is a fit to $\sim L^{2\zeta}$ with $\zeta = 2/3$, suggesting 1D RB disorder. Inset: PFM phase image of the stripe domains. (d) Device schematic of PZT top-gated MoS_2 transistor.

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Epitaxial SrTiO₃ Films with Dielectric Constants Exceeding 25,000

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SrTiO₃ (STO) is an incipient ferroelectric perovskite oxide for which the onset of ferroelectric order is suppressed by quantum fluctuations. This property results in a very large increase in static dielectric constant from ~300 at room temperature to ~20,000 at liquid He temperature in bulk single crystals. However, the low-temperature dielectric constant of epitaxial STO films is typically a few hundred to a few thousand. In this talk, we will resolve this long standing issue.

Using all-epitaxial capacitor structure of the form *n*-STO/undoped STO/*n*-STO (001) prepared by hybrid molecular beam epitaxy, we demonstrate intrinsic dielectric constants of an unstrained STO (001) film exceeding 25,000 at low temperature. A careful analysis of the temperature-dependent dielectric constants reveal that the *n*-STO/undoped STO interface plays a vital role in determining the dielectric properties. Furthermore, using different dopant-types in *n*-STO, we reveal that it is the depleted side of the interface, i.e. *n*-STO that governs the overall measured dielectric constant [1]. A detailed growth study combined with the temperature- and frequency-dependent dielectric measurement will be presented.

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High-Mobility Field-Effect Transistor Using a 2-Dimensional Electron Gas at the LaScO₃/BaSnO₃ Interface

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A 2-dimensional electron gas (2DEG) system with high mobility was discovered at the interface of two perovskite oxides: a polar orthorhombic perovskite LaScO₃ and a nonpolar cubic perovskite BaSnO₃. Upon depositing the LaScO₃ film on the BaSnO₃ film, we measured the conductance enhancement and the resulting 2DEG density (n_{2D}). Comparing the results with the previously reported LaInO₃/BaSnO₃ polar interface [1,2], we applied the “interface polarization” model to the LaScO₃/BaSnO₃ system, in which the polarization exists only over four pseudocubic unit cells in LaScO₃ from the interface and vanishes afterward like the LaInO₃/BaSnO₃ interface. Based on the calculations of the self-consistent Poisson–Schrödinger equations, the LaScO₃ thickness dependence of n_{2D} of the LaScO₃/BaSnO₃ heterointerface is consistent with this model. Furthermore, a single subband in the quantum well is predicted. By use of the conductive interface and the LaScO₃ as a gate dielectric, a 2DEG transistor composed of only perovskite oxides with high field-effect mobility (μ_{FE}) close to $100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is demonstrated. [3]

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Band-Engineered LaFeO₃-LaNiO₃ Interfaces for Electrocatalysis of Water

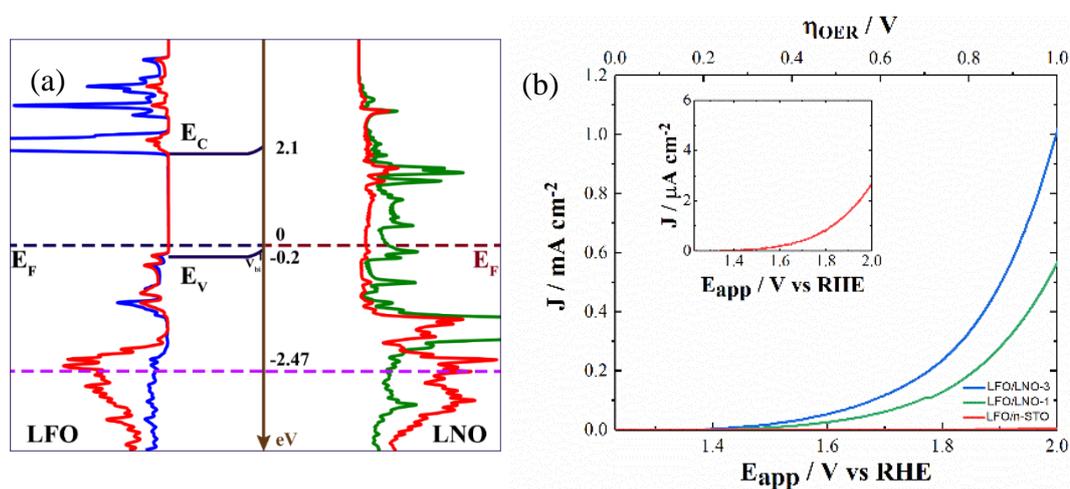
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Perovskite oxide thin films have been studied extensively for applications in electrocatalysis of water. Particularly, LaFeO₃ and LaNiO₃ have shown great potential as catalysts in the oxygen evolution reaction. However, their catalytic performance needs to be improved significantly for commercial applications to compete with precious metal catalysts. Through band engineering we can fine tune the electronic properties of the material that might lead to the efficient energy conversion. By choosing suitable materials, charge can be driven across the heterostructure interface, which often leads to enhanced catalytic performance. The band alignment at the interface is crucial for charge transfer. In this work, we investigated the band offset at the LaFeO₃/LaNiO₃ interface combining experimental techniques as well as first principles calculations.

LaFeO₃/LaNiO₃ heterostructures were synthesized using molecular beam epitaxy. We performed band offset calculation from XPS data. Our result shows that charge transfer at the interface is minimal but that the valence band offset between the top LaFeO₃ layer, and the metallic LaNiO₃ Fermi level is only ~0.2 eV, pinning the LaFeO₃ in a p-type state. Models of the heterostructures using first-principles calculations within the density functional theory support this conclusion. Electrochemical measurements produced on these heterostructures revealed enhanced catalytic performance with respect to individual thin films. Comparison of these results with LaFeO₃ thin films indicate that thickness as well as doping between *n*-type, intrinsic, and *p*-type all play a role in the catalysis that may be exploited to further improve the efficiency of oxygen evolution reactions [1,2].



Figure

- (a) Experimentally-validated band alignment from density functional theory modeling of LFO/LNO interface; (b) Enhanced catalytic performance of LFO/LNO heterostructure compared to equivalent-thickness LFO film on Nb-doped SrTiO₃

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A Laser-ARPES View of the 2D Electron Systems at LaAlO₃/SrTiO₃ and Al/SrTiO₃ Interfaces

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The electronic structure of the two-dimensional electron system (2DES) found at the Al/SrTiO₃ (Al/STO) and LaAlO₃/SrTiO₃ (LAO/STO) interfaces is measured by means of laser angle resolved photoemission spectroscopy (ARPES), taking advantage of the large photoelectron escape depth at low photon energy to probe these buried interfaces. The possibility of tuning the electronic density in Al/STO by varying the Al layer thickness is demonstrated, [1] paving the way for a designed band structure in devices. [2] A new open source Python code released under the name *BinPo* is used to compute the electronic structure of the 2DES. [3] We show that the electronic structure evolution for different densities differs qualitatively from a rigid band shift model. A comparison of the ARPES measurements in LAO/STO and Al/SrTiO₃ allows us to show that both 2DES are strongly coupled to longitudinal optical phonons, in agreement with previous reports of a polaronic ground state in similar STO based 2DESs.[4] Tuning the electronic density in Al/STO to match that of LAO/STO and comparing both systems, it is estimated that the intrinsic LAO/STO 2DES has a bare band width of ≈ 60 meV and a carrier density of $\approx 6 \times 10^{13} \text{ cm}^{-2}$. [1]

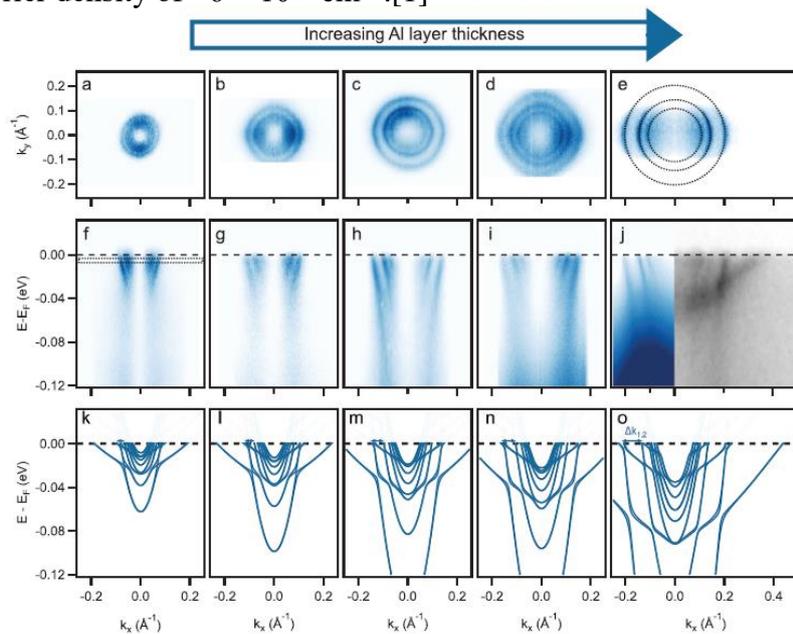


Figure 1. (a-e) ARPES Fermi surface of the 2D electron system stabilized at the Al/SrTiO₃ interface for increasing Al thickness. These plots are obtained by summing two measurements with s and p polarization, respectively, and integrating the spectra within an energy window of ± 5 meV around E_F . f–j) Energy–momentum dispersion measured along the [100] high symmetry direction ($k_y = 0$). The right half of panel (j) shows synchrotron data acquired in the second Brillouin zone with a photon energy of 52 eV on a bare STO surface cleaved in UHV. All other data on Al/STO were acquired in the first Brillouin zone with $h\nu = 6$ eV. (k–o) Self-consistent tight binding supercell calculation of the electronic band structure. The electronic densities are from first to last column: 6, 10, 14, 17, and 31 (10^{13} cm^{-2}).

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Tunable spin and orbital Edelstein effect at (111) LaAlO₃/SrTiO₃ interface

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Converting charge current into spin current is one of the main mechanisms exploited in spintronics [1]. One prominent example is the Edelstein effect [2], namely the generation of a magnetization in response to an external electric field, which can be realized in systems with lack of inversion symmetry. If a system has electrons with an orbital angular momentum character, an orbital magnetization can be generated by the applied electric field giving rise to the so-called orbital Edelstein effect [3]. Oxide heterostructures are the ideal platform for these effects due to the strong spin-orbit coupling and the lack of inversion symmetries [4-5]. Beyond a gate-tunable spin Edelstein effect, we predict an orbital Edelstein effect an order of magnitude larger than the spin one at the (111) LaAlO₃/SrTiO₃ interface. We model the material as a bilayer of t_{2g} orbitals using a tight-binding approach, while transport properties are obtained in the Boltzmann approach. We give an effective model at low filling which explains the non-trivial behaviour of the Edelstein response, showing that the hybridization between the electronic bands crucially impacts the Edelstein susceptibility.

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Controlling the order parameter coupling in nickelate based superlattices

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Rare-earth nickelates (chemical formula $RNiO_3$, R = rare earth) belong to the family of transition metal perovskites. These materials display a characteristic temperature-dependent metal-to-insulator transition (MIT) together with a lowering of the crystal symmetry from the high temperature orthorhombic metallic phase (Pbnm symmetry) to the low temperature monoclinic insulating phase ($P2_1/n$ symmetry), concomitant with a breathing distortion, or bond disproportionation, of the NiO_6 octahedra units [1,2].

Many studies have focused on manipulating the MIT for instance by using strain [3], changing growth direction [4] or designing heterostructures [5].

Using nickelate superlattices we recently studied the characteristic length scale over which a metallic or an insulating phase can be established and the physics that sets this length scale [6]. By growing $SmNiO_3/NdNiO_3$ superlattices and using several experimental and theoretical methods we demonstrated that the length scale mainly depends on the interplay between the energy cost of the boundary between a metallic and insulating region and the energy gain of the bulk phases [6]. Additionally, in samples displaying phase coexistence, room temperature STEM-EELS (Scanning Transmission Electron Microscopy - Electron Energy-Loss Spectroscopy) shows that the phase boundary is very sharp, of the order of a few unit cells [7].

To further understand this unusual coupling and the evolution of the structural and electronic order parameters across these phase boundaries, $SmNiO_3/NdNiO_3$ superlattices were grown with additional $LaAlO_3$ insulating spacer layers. The samples are grown using RF off axis magnetron sputtering, and are morphologically and structurally characterized by atomic force microscopy (AFM) and X-ray diffraction (XRD) respectively. Temperature dependent transport measurements as a function of the thickness of the spacer will be presented, and should give a first indication of how the metal-to-insulator transitions couple in this novel superlattice system.

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Properties of the spin-polarized 2DEG at the LAO/ETO/STO interface

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The multi-orbital character of quasi-two-dimensional electron gases (q2DEGs) at the interface between oxide materials is at the origin of the rich physics of these systems and of the emergence of unconventional quantum phases in them. We will present the two-dimensional electron system (2DES) at the LaAlO₃/EuTiO₃/SrTiO₃ interface. It shows tunable superconductivity, spin-orbit coupling and ferromagnetism [1], offering the possibility to study the interaction among ground states that are rarely found combined in the same system, and opening to the possibility to interesting electronic applications. We explore the nature and properties of LAO/ETO/STO interface using a wide range of techniques, from electrical transport to angle-resolved photoemission spectroscopy. We will compare the experimental results with Density functional theory calculations, resolving the mechanism of orbital selective switching of the spin-polarization in this oxide q2DEG [2]. The joint experimental and theoretical approach is becoming essential to gain detailed understanding of novel heterostructures showing exceptional properties, and as a guide for future design of materials for advanced applications.

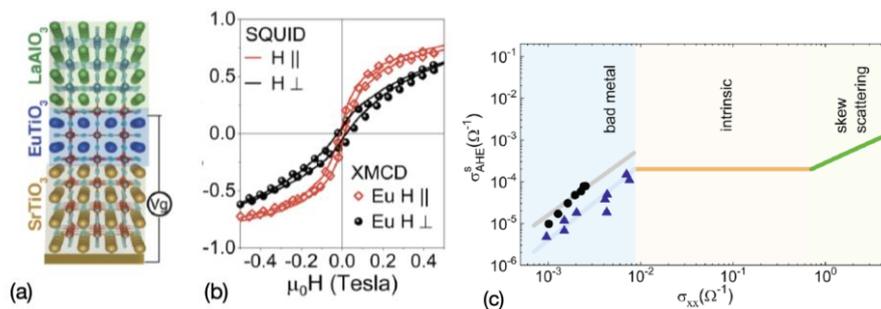


Figure 1: (a) Sketch of the LAO/ETO/STO heterostructure. (b) Eu- XMCD (scatter data) and SQUID magnetization (continuous lines) as function of the magnetic field parallel (red) and perpendicular (black) to the interface. The data are normalized to the saturation value. (c) Anomalous Hall conductivity vs. longitudinal 2D conductivity. Full lines refer to the exponent α in the relation $\sigma_{\text{AHE}} \propto (\sigma_{\text{xx}})^\alpha$: $\alpha = 1.8$ (light blue, and gray, intrinsic anomalous Hall effect suppressed by disorder), $\alpha = 0$ (orange, intrinsic anomalous Hall effect), $\alpha = 1$ (green, skew scattering).

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Light-enhanced gating effect at the conducting EuO-KTaO₃ interface

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Light illumination and electrostatic gating field are two widely used stimuli for controlling electronic processes in low dimension systems. KTaO₃(KTO)-based conducting interfaces have gained tremendous interest because its spin-orbit coupling strength is one order of magnitude higher than STO [1], which makes it a promising candidate for spintronics and optoelectronic devices. In this talk, I will present the combined effect of light illumination and electrostatic gate on the conducting EuO-KTO interface. An unusual illumination-enhanced gating effect (three order change in conductance) is observed for this metallic system at room temperature. This enormous change in conductance is observed even with visible light of very low power intensity of 0.5 mW along with the back gate. This unusual effect offers a new perspective for tuning the photoelectrical properties at the oxide interfaces, which can be helpful for designing advanced photoelectric devices with high-performance and multifunctionality.

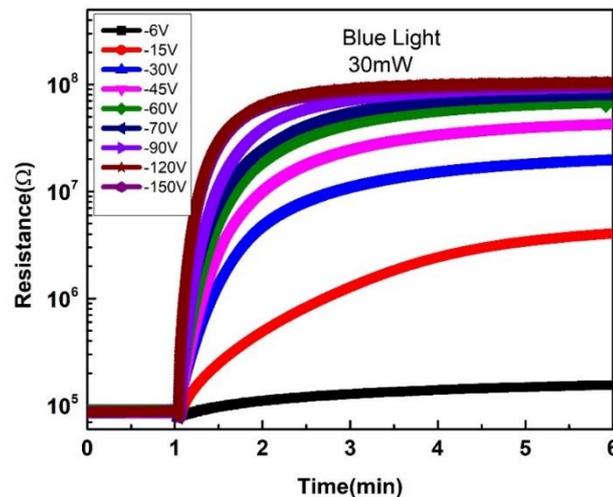


Figure 1: Combined effect of the negative electrostatic gate and light (405 nm) illumination of 30 mW power intensity on the conducting EuO-KTO interface.

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Frustrated Magnetism in Rare-earth Titanate Pyrochlore Thin Films Grown by Molecular Beam Epitaxy

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Frustrated magnetism generated by the network of corner-sharing tetrahedra in pyrochlores leads to exotic magnetic behavior ranging from unusual ordered phases to quantum spin ice and quantum spin liquid ground states. In thin films, the interactions between magnetic cations can be tuned with epitaxial strain, dilution with non-magnetic ions, and varying film thickness. Thus, the thin film geometry provides fertile ground to manipulate and probe the unusual magnetic properties of pyrochlores in ways that are inaccessible in bulk crystals. Using reactive-oxide molecular beam epitaxy, we have produced thin films of (111)-oriented $\text{Tb}_2\text{Ti}_2\text{O}_7$ with high crystalline quality confirmed by in-situ RHEED, x-ray diffraction, x-ray absorption spectroscopy, and atomic-resolution STEM. We use SQUID magnetometry to study the thickness dependence of magnetic behavior in pure $\text{Tb}_2\text{Ti}_2\text{O}_7$ films and heterostructures with non-magnetic, isostructural $\text{Y}_2\text{Ti}_2\text{O}_7$ layers.

Evidence of interfacial Mn-ion displacement and lattice modulations coupled magnetic properties in LSMO films

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The properties of half-metallic manganite thin films depend on the composition and structure in the atomic scale, and their potential functional behavior can only be understood by precise structure characterization. By combining advanced transmission electron microscopy, electron energy loss spectroscopy, we provide an evidence of a 7 nm-thick interface layer in La_{0.7}Sr_{0.3}MnO₃ (LSMO) thin films, always termed as dead layer, attributed to an elongation of out-of-plane lattice parameter and Mn valence states etc. Surprisingly, along with previous observations, we, for the first time observed a structural shift of Mn ions coupled with oxygen vacancies and a reduced Mn valence state within dead layer [1].

We will also present an interesting observation of the magnetic properties as a function of LSMO film thickness. The *in-plane* magnetic anisotropy symmetry landscapes observed by MOKE found to be strongly coupled with the *in-plane* lattice modulations / artificial superlattices formed by accommodating the epitaxial strain in the form of tilting of the unit-cell i.e., shear strain [2, 3]. Such lattice modulations are dominant enough to overcome the strong magnetocrystalline anisotropy, often observed at low temperatures. Our findings provide future directions to understand and control the lattice modulations in the oxide heterostructures and could open a pathway tailoring functional properties into device applications.

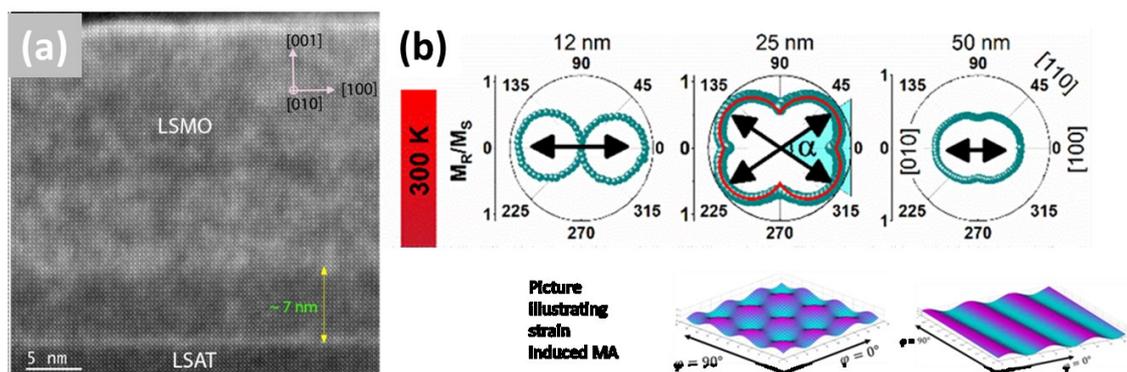


Figure: a) STEM–ADF image showing the contrast variation at around 7 nm distance from the interface, and (b) Thickness-dependent magnetic anisotropic polar plots and its associated lattice modulations.

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Proximity induced ferromagnetic state in the spin-orbit semimetal SrIrO₃

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The 5*d* iridium-based transition metal oxides (TMOs) of the Ruddlesden-Popper series Sr_{*n*+1}Ir_{*n*}O_{3*n*+1} have gained huge and broad interest because of the strong spin-orbit coupling in these compounds favoring new or exotic quantum states. This has resulted in one of the most active and competitive front of condensed matter research.

In contrast to the 3*d* TMOs, the electron-electron correlation strength is however generally too small in the 5*d* TMOs to yield *e.g.* ferromagnetism, limiting the field of practical applications of these materials.

Recently, we succeeded to produce a ferromagnetic (FM) state in the spin-orbit semimetal SrIrO₃ (SIO) by proximity effect in contact with an FM insulator, LaCoO₃ (LCO) [1]. The FM state of SIO was selectively probed by electrical transport in SIO/LCO thin film heterostructures. A large intrinsic and positive anomalous Hall effect (AHE) appears for $T \leq 100$ K and indicates magnetic ordering. The AHE shows unusually large coercivity below about 40 K, very likely caused by a strong pseudo-spin lattice coupling. The anisotropic magnetoresistance (AMR) displays four-fold magneto-crystalline anisotropy below T_C with in-plane $\langle 110 \rangle_{pc}$ magnetic easy axis.

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Application of Rashba-Edelstein effect in ferromagnet/2DEG based Spin-to-charge interconverters

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The Magnetolectric Spin-Orbit (MESO) technology [1] is a relatively new application coming from the Spin Orbitronics field. MESO devices combine the advantages of Spin Orbit coupling (SOC) (read out part) and the Magnetolectric effect (input of the device), to achieve an efficient Spin-to-charge conversion (SCC) and the capability of storing non-volatile information in the magnetic state of the device. In this work, we focus on the MESO device's read-out part, using a 2-dimensional electron gas (2DEG) as a high SOC material [2]. The two principal advantages of using 2DEGs are the finite Rashba SOC they present, and the high tunability of their electronic structure, which is related to the Rashba-Edelstein effect (IREE) response [3]. This section consists of applying a charge current through a ferromagnetic material, and thus, generating a spin current that will flow from the ferromagnet into the high SOC material. Due to the presence of inverse IREE, which naturally appears in 2DEGs [4,5], the spin current will be transformed into a perpendicular charge current that will carry the information about the magnetization of the ferromagnet. In our study, we have designed, patterned and fabricated T-shaped nanodevices where the charge current is injected into a Co layer (the ferromagnet), and there it is transformed into a spin current that will flow into the 2DEG, generating a perpendicular voltage output. Interestingly, we found signatures of SCC and studied its evolution as a function of the temperature while applying back-gate, magnetic-field or rotation. Furthermore, since the current injected into the 2DEG is not a pure spin current, but a mixture of spin and charge currents (i.e. a spin-polarized current), we tried to disentangle the IREE contribution from different spurious effects like Planar Hall Effect, Anomalous Hall Effect and anisotropic magnetoresistance. Our findings are very promising in terms of spin-logic approaches for alternative applications in computing field, paving the way towards a new generation of scalable, non-volatile and high energy efficiency beyond-CMOS spintronic logic devices.

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Correlating thickness and temperature dependent structural, electronic and magnetic transitions in epitaxial SrRuO₃ thin films

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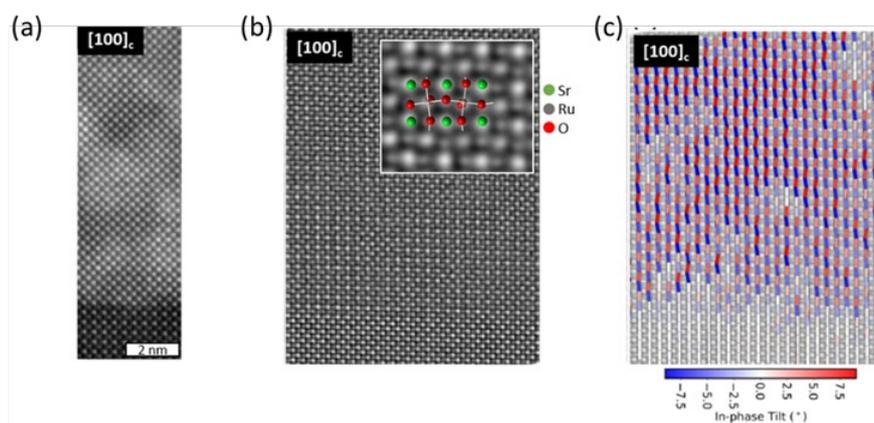
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SrRuO₃(SRO) is a 4d itinerant ferromagnetic metal which is widely used as an oxide electrode in all-oxide electronic devices. Epitaxial thin films of SRO have been shown to exhibit interesting physical properties including strain and thickness-dependent metal-insulator and magnetic transitions and signatures of the topological Hall effect.[1,2] To understand the origin of the thickness-dependent properties of the SRO system, we investigate the temperature-dependent layer-resolved structure of 3 to 44 unit cell thick SRO films using a combination of high-resolution synchrotron X-ray scattering measurements and transmission electron microscopy.[3] The oxygen octahedral tilts and rotations and Sr displacements characteristic of the bulk orthorhombic phase of SRO are found to be strongly dependent on temperature, the film thickness, and the distance away from the film–substrate interface. The orthorhombic distortions related to the transport and magnetic properties of SRO are found to be suppressed in the 2–3 interfacial layers due to structural coupling with the SrTiO₃ substrate and correlate with the critical thickness for ferromagnetism in uncapped SRO films.



Layer-resolved structure of a 44 unit-cell SRO film. (a) Annular dark field images of the 44 uc SRO film along the [100]_c projections with (b) the corresponding integrated differential phase contrast images. The inset shows a magnification of the iDPC image with the XRD structure overlaid. (c) Measured in-phase tilting angle map along the [100] zone axis.

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Coupling of octahedra rotations in orthorhombic perovskite heterostructures

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Octahedral rotations patterns in transition metal perovskites affect their physical properties due to the strong coupling between the lattice and the electronic degrees of freedom. Across an interface in an epitaxial heterostructure, the corner sharing of the oxygen octahedra couples the layer-substrate or layer-layer octahedral rotation patterns in competition/together with the epitaxial strain. The consequences and the extent of the rotations imprint are currently at the focus of several studies, since theoretical work indicates that it could be at the origin of novel properties and functionalities.

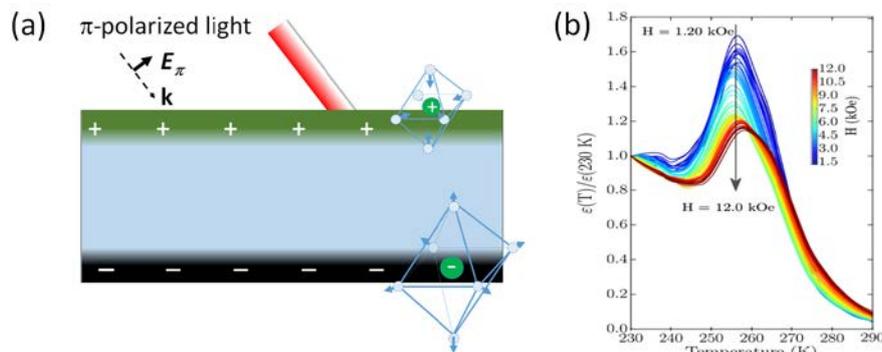
This work investigates the microscopic details of orthorhombic perovskite heterostructures using X-ray diffraction, scanning transmission electron microscopy (STEM) and second-principles simulations, revealing that the interface lattice results from a delicate balance between elastic strain energy and rotation modification energy. We observe that the substrate imprints a rotation pattern into thin layers despite the induced elastic strain cost; for thicker films, a transition layer accommodates the change in the rotation pattern between the substrate and the bulk of the layer. We experimentally probe this effect in LaVO₃ thin films epitaxially grown by pulsed laser deposition onto DyScO₃ substrates and theoretically confirm this behavior in CaTiO₃ structures. This phenomenon opens the way to the control of the orientation of the crystalline axes which often correspond to easy magnetization axes or polarization directions.

Light-matter interactions modulated by electron-lattice coupling in transition metal oxides

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The coupling of electrons to the lattice is important in condensed matter physics, particularly in transition metal oxides where narrow d -bands promote electronic correlations while enhancing electron-phonon coupling. Here we show that modulating the strength of this coupling offers the opportunity to tailor materials properties. We first discuss the case of transparent conductive SrVO_3 , for which we recently found that electron-phonon coupling plays a pivotal role on the observed enhancement of the carrier effective mass [1]. In contrast to the conventional view, our study establishes that polaronic transport, rather than e - e correlations, drives the mass enhancement and is responsible for transport and optical properties. Resulting from this mass enhancement, the plasma frequency is pushed down to the near-infrared region. Plasmon resonances in unpatterned SrVO_3 films are excited by exploiting charge density gradients at the surface, at frequencies determined by the effective mass [2], see Figure 1a. Our observation points to the existence of polaronic plasmon excitations in heterostructures containing 3d and 4d metallic oxides, offering new perspectives for photonics and optoelectronics. On the other hand, in materials with narrower bandwidth, localized polarons rather than itinerant polarons come to stage. We discuss light-matter interactions in $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, where we observe large magneto-optic responses around the Curie temperature at a certain range of frequencies (Figure 1b). We show that such responses are related to spin-orbit coupling and Jahn-Teller instabilities [3, 4]. As a result, at resonant frequencies, light induces transitions where the spin is inverted, which is detected by circularly polarized light. Such interactions offer the opportunity to use light to entangle orbital and spin degrees of freedom, and explore nontrivial quantum phenomena in 4d/5d quantum systems coupled to electromagnetic fields.



(a) Sketch of light coupling to plasmon-polarons, as in the case of SrVO_3 films (“+” and “-” indicate excess of charge).
(b) Temperature-dependence ellipticity (ϵ) of $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ at $\lambda = 402$ nm for different magnetic fields. [3,4].

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Deep-UV transparent conducting oxide La-doped SrSnO₃ with high figure of merit

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Perovskite stannate SrSnO₃ (SSO) is attracting attention as ultraviolet transparent conducting oxide (UV TCO) due to its ultra-wide band gap and high conductivity. Here, we investigate in detail the thickness-dependent electrical, structural and optical properties of sequentially strain-relaxed La-doped SrSnO₃ (SLSO) epitaxial thin films. We find the SLSO films grow as an orthorhombic *Pnma* phase with $a^-a^-c^+$ in the c^+ direction under the tensile strain. With the strain relaxation as the films become thicker, vertical grain boundaries are created and the orthorhombic phase becomes reoriented to all three possible orientations. Simultaneously, the conductance starts to deviate from the linear behavior with increasing the film thickness. Through the analyses of thickness fringes in optical transmittance we found that 120 nm thick nominally 4% La-doped SrSnO₃ film has a figure of merit ($\phi_{TC} = 2.65 \times 10^{-3} \Omega^{-1}$) at $\lambda = 300$ nm in the deep-UV region, which is the highest value among the well-known candidates for UV TCOs reported to date.

Coexistence of floxo-field and metallicity near crack in SrRuO₃ thin films

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Strain engineering is one of the typical and powerful methods to manipulate correlations between degrees of freedom, such as lattice, spin, and charge. In this paradigm, basically, coherent growth of a thin film has been recognized as the first priority to explore the artificial nature. Recently, nanoscale crystal defect created by strain relaxation has been introduced as a new route to access novel structure, and highly advanced local probing techniques have realized observation of defect states and their physical properties. Here, we investigate local properties near crack in SrRuO₃ thin films grown on KTiO₃ substrate. The structure and its properties near cracks are observed by adopting local probing techniques. As the film thickness increases, the fully clamped crystal lattice relaxes and nanoscale line crack occurs above 15 uc. From the scanning transmission electron microscopy result, we found that spatial size and the number of cracks increase, and a huge strain gradient with an order of 10^7m^{-1} is realized near the crack. Moreover, we monitor the space-resolved optical response near crack using nanoscale infrared spectroscopy. The observed local conductivity increases near crack due to lateral strain relaxation. Based on the experimental results, we discuss the feasibility to realize a highly multifunctional phase by crystal defect.

Electronic states and octahedral tilting in SrNbO₃

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Octahedral tilting is a common feature of perovskites and is relevant for many physical phenomena, ranging from electronic and magnetic properties to metal-insulator transitions. Knowledge about how different tilting modes can be achieved by external stimuli is of great importance. [1] Among a wide range of 4d perovskite oxides, the effect of octahedral tilting on the intrinsic electronic properties of SrNbO₃ is still not thoroughly explored.

Here we aim at revealing and understanding the interplay between strain and octahedral tilting in the perovskite SrNbO₃ using density functional theory (DFT). SrNbO₃ resembles SrTiO₃, but it has a larger lattice parameter and partially occupied *d* states (*4d¹*). Energy favorable tilts are found for SrNbO₃ as a function of biaxial strain. The coupling between the tilts and electronic states is studied by investigating the electronic band structures. The analysis of the band structures is twofold: (1) the band structures of the primitive cells are analyzed and then (2) the bands of supercells are unfolded to make for easier comparison with angle-resolved photoemission experiments. Unfolded Fermi slices are also investigated where the changes and additional states due to tilting is visible. We also report how the states vary with the octahedral tilt, which can be tuned by biaxial strain. SrNbO₃ exhibits two tilt modes with similar energy under compressive biaxial strain. The out-of-phase tilt shows gapless linear dispersion near the Fermi level [2] while, interestingly, according to our results the in-phase tilt shows a tunable gapped linear dispersion near the Fermi level.

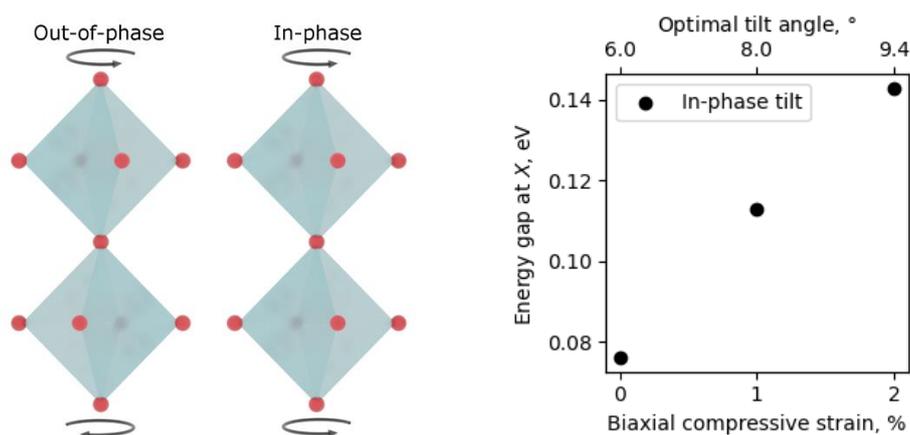


Figure: (Left) Comparison of out-of-phase and in-phase octahedral tilting. For simplicity the A and B ions are not shown. (Right) Demonstration of the tunability of the gapped states for the in-phase octahedral tilt mode. Note, the gap is only in vicinity of the X symmetry point, i.e. there are available states at other points in k-space.

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Free-standing SrMoO₃ single-crystal like thick films

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Recently, the growth of 5 μm thick fully strained epitaxial SrMoO₃ films on GdScO₃ substrates was reported by our group [1]. It is remarkable, that coherently strained growth can be maintained in such thick perovskite films going considerably beyond typical critical thicknesses of perovskite thin films with 0.2% compressive lattice mismatch. This study aims at (i) identifying the contributing mechanisms to the large critical thickness and (ii) exploring the feasibility of wafer transfer by investigating free-standing SrMoO₃ films grown by pulsed laser deposition (PLD) onto X ScO₃ (X = Dy, Tb, Gd, Sm) substrates using a water-soluble sacrificial layer of Sr₃Al₂O₆. The sacrificial layer and SrMoO₃ films adopt the in-plane lattice parameters of the substrates ranging from 3.950 Å (DyScO₃) to 3.989 Å (SmScO₃). Free-standing SrMoO₃ films were produced by dissolving the sacrificial layer in water and transferring the SrMoO₃ layer to Si substrates, following the recipe reported in the pioneering work of D. Lu *et al.* [2]. The lattice parameters of the epitaxially strained and freestanding SrMoO₃ films were measured before and after the lift-off, respectively, using X-ray diffraction (XRD). The Sr/Mo ratio in the films was calculated from X-ray photoelectron spectra of the films measured before the lift-off.

The dependence of the lattice parameters and Sr/Mo cation ratio on the in-plane strain is shown in Fig. 1 (a) and (b). The lattice parameter *c* of the strained films corresponds to that calculated with a computed Poisson's ratio of 0.271 [3]. Consistent linear changes of the Sr/Mo ratio are observed for the compressively strained SrMoO₃ on DyScO₃ (-0.8%) to the tensile strained SrMoO₃ on SmScO₃ (0.3%), respectively. The four key results are: (i) The main contribution to the overall strain comes from the biepitaxial strain of the substrate. (ii) Increasing in-plane lattice spacings of SrMoO₃ result in an increased Sr excess in SrMoO₃. (iii) Wafer transfer is possible despite the large thickness of the layers. (iv) The μm thickness together with the macroscopic lateral dimensions allow the classification of the free-standing SrMoO₃ thick films as novel type of single crystals.

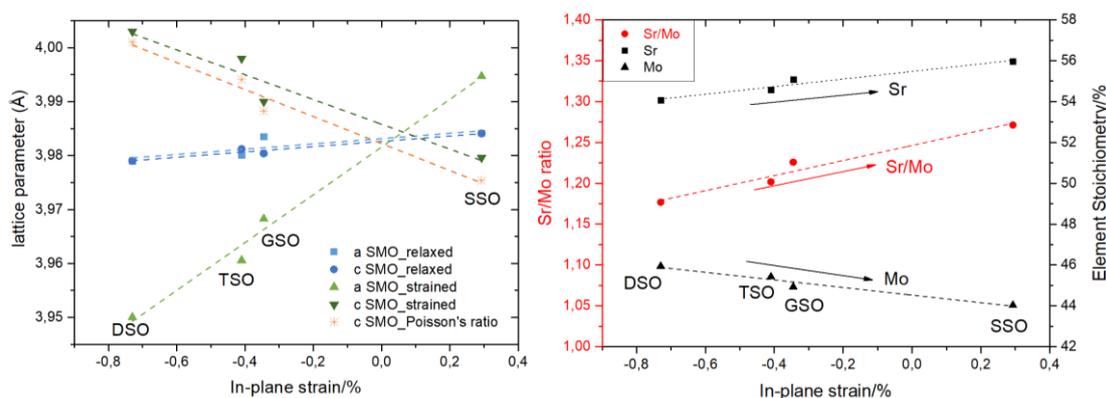


Figure 1 (a) Lattice parameters and (b) Sr/Mo cation ratio of the epitaxially grown strained and free standing SrMoO₃ films as functions of the in-plane strain.

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Structural and Optical Properties of Erbium-doped Anatase TiO₂ Thin Films Grown by Molecular Beam Epitaxy

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Rare-earth ions (REI) doped into solid-state crystal hosts offer an attractive platform for quantum information processing due to long coherence times and potential scalability. Recent efforts to increase coherence times in REI defects have focused on finding suitable host crystals with minimal sources of decoherence (e.g., nuclear spin, oxygen vacancies, and strain variation). In this respect, molecular beam epitaxy (MBE) offers opportunities, as it can grow epitaxial films using highly pure elemental sources. In addition, MBE growth can provide these materials in thin-film and heterostructure forms that provide a flexible host crystal environment, enabling scalable on-chip integration with other quantum devices. Here, we report on the MBE growth of Er-doped anatase TiO₂ thin films on LaAlO₃ (001) substrates with varying doping concentrations. Their structural and optical properties are characterized using x-ray diffraction and photoluminescence (PL) measurements to investigate how factors such as the Er concentration, buffer layer, and delta-doping affect the optical inhomogeneous linewidths of Er³⁺ ions. By adjusting Er-dopant concentration, we achieve optical inhomogeneous linewidths of 5 GHz at 4.5 K. We show that 9 nm-thick buffer and capping layers can further reduce the linewidth by 40%, suggesting a pathway to further narrowing linewidths in this system.

Large-scale Screw Dislocation Grids in Oxide Heteroepitaxies

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Screw dislocation is still a mystery in material physics since the proposal and corroboration of dislocation linear defects for understanding properties of materials. While screw dislocation is important not only for understanding plastic deformation of crystals such as cross slipping[1], but also for optical and electrical properties of materials[2], the structures and related experimental studies on screw dislocations are still less known since there is almost no atom distortion when viewed along dislocation line. This fact makes great difficulties for searching and studying screw dislocation related material responses. For instance, one cannot even aware the existence of a screw dislocation in an atomic scale high resolution image even there are many screw dislocations. In particular, it is theoretically known that shear strains between twist grain boundaries and heteroepitaxy systems may be relaxed via screw dislocation grids. However, the specific structure and the thickness depended evolutions of these screw dislocation grids are still largely unknown, especially for oxide films. Here, by using orthorhombic (001)-oriented DyScO₃ exhibiting potentially large shear strain as substrates, we have directly observed the large-scale screw dislocation grids in this DyScO₃/BiFeO₃ oxide heteroepitaxies. Pure screw dislocations with $a[100]$ and $a[0\bar{1}0]$ Burgers vectors were confirmed by multi-scale transmission electron microscopy study. Specifically, a shear strained transition layer of several unit cells thickness was obtained near the interface. Our results suggest a practical platform for manipulating and studying structures and induced responses corresponding to screw dislocations. Moreover, these results directly confirm screw dislocation grids as a factor to tailor shear strains in epitaxial systems, which will be helpful for guiding strain engineering for oxide heteroepitaxies via shear strains.

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Limits to the strain engineering of layered square-planar nickelate thin films

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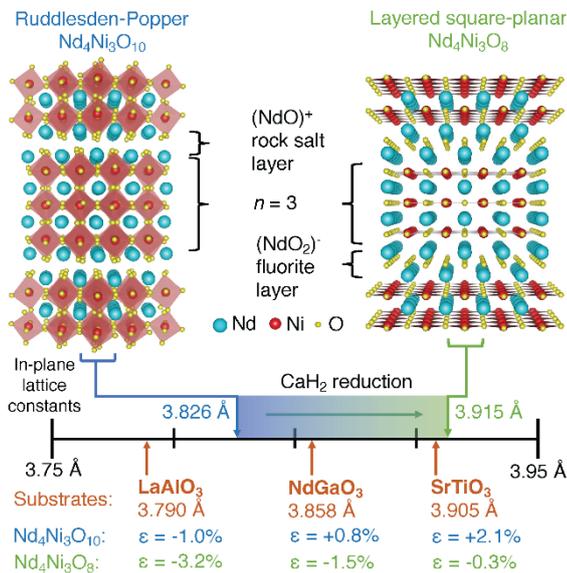
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Since the discovery of superconductivity in $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$, square-planar nickelates have become a prime testbed to study superconductivity in a cuprate-like materials analogue [1]. The layered square-planar nickelates, $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$, are an appealing system to further tune the electronic properties by dimensionality, with superconductivity recently observed in thin film $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ [2]. Notably, the layered $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$ nickelates and their parent $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ oxygenated forms have yet to be synthesized in bulk for $n>3$. Here, we use molecular beam epitaxy and subsequent CaH_2 reduction to study the stability of layered square-planar nickelate thin films under different strain states on LaAlO_3



Schematic crystal structures of (left) $\text{Nd}_4\text{Ni}_3\text{O}_{10}$ and (right) $\text{Nd}_4\text{Ni}_3\text{O}_8$. The number line presents the bulk in-plane lattice parameters of $\text{Nd}_4\text{Ni}_3\text{O}_{10}$ and $\text{Nd}_4\text{Ni}_3\text{O}_8$ as well as the pseudocubic lattice parameters of LaAlO_3 (100), NdGaO_3 (110), SrTiO_3 (001).

(100), NdGaO_3 (110), and SrTiO_3 (001). We explore the competing growth requirements for the epitaxial stabilization of the parent $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ compounds and subsequent reduction to $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$. The highest quality precursor $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ films are synthesized on LaAlO_3 under compressive strain, while films under tensile strain on NdGaO_3 and SrTiO_3 form strain relieving vertical rock salt faults. Due to the $\sim 3\%$ expansion of the in-plane lattice constant upon reduction, however, a high-quality metallic reduced phase can be achieved only on NdGaO_3 , which limits the compressive strain in the reduced state to 1.5%. In contrast, layered square-planar nickelates on LaAlO_3 , which provides 3.2% compressive strain, are insulating despite the presence of large pristine regions of the square-planar phase. Our work not only provides a pathway to the stabilization of square-planar $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$ thin films but further benchmarks the limits to strain engineering these materials.

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Intrinsic magnetism in superconducting infinite-layer nickelates

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The discovery of superconductivity in Nd_{0.8}Sr_{0.2}NiO₂ [1] introduced a new type of layered superconductor that is now known to be a part of a growing family of nickelate superconductors.

In the infinite-layer nickelate system, RNiO₂, X-ray scattering work has uncovered magnetic excitations that persist across the superconducting dome [2], while neutron scattering on bulk powder samples of the parent compound suggests no long-range magnetic order [3]. This already hints at a difference between the superconducting nickelates and the superconducting cuprates, in which the parent compounds are long-range ordered antiferromagnets and the order is quenched with hole doping [4].

In order to gain further insight into the magnetic character of these compounds, we have carried out low energy muon spin rotation/relaxation studies on a series of infinite-layer nickelate thin films with $R = (\text{La}, \text{La}_{0.8}\text{Sr}_{0.2}, \text{Pr}_{0.8}\text{Sr}_{0.2} \text{ and } \text{Nd}_{0.825}\text{Sr}_{0.175})$ [5]. Regardless of the rare earth ion or doping, we find a magnetic ground state that is likely to be short-range ordered and antiferromagnetic, arising from local moments on the nickel sublattice. Weak transverse field measurements as a function of temperature and implantation energy show that the nickelate layer is the magnetic component in the heterostructure and has 100% magnetic volume fraction. Crucially, this behaviour starts to appear at around 150 K and holds down to the lowest temperature measured, which is within the superconducting state.

The coexistence of magnetism with superconductivity, as well as the high temperature onset, highlights the nickelates as qualitatively distinct from the doped cuprates. These newly-discovered superconductors may be more reminiscent of some iron pnictides and heavy fermion compounds. This may be as a result of their suspected multi-orbital nature.

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Enhancing superconductivity in high T_c cuprates via high-frequency phonons

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Cuprates, Mott insulators in their normal states, are known to host unconventional high-temperature superconductivity [1,2], whose mechanism is still under debate [3,4,5]. Light elements such as hydrogen and lithium lead to high-frequency phonons, which can give rise to high-temperature superconductivity within the BCS theory framework [6,7,8]. Despite the lack of clarity regarding the superconducting mechanism in cuprates, one may expect to see enhanced electron-phonon coupling and hence enhanced superconductivity by introducing high-frequency phonons into cuprates. In this first principles theoretical study, we choose $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_8$ (BSCCO) as a prototype cuprate, and use density functional theory methods to study its phonon and superconducting properties under the effects of (1) calcium-to-lithium substitution, and (2) lithium-dopant-assisted hydrogen intercalation. Synthesis and characterization of such light-elements-doped BSCCO are concurrently carried out by our experimental collaborators.

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Superconductivity in a quintuple-layer square-planar nickelate: superconducting and electronic properties

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Since the discovery of high-temperature superconductivity in copper oxide materials, there have been sustained efforts to both understand the origins of this phase and discover new cuprate-like superconducting materials. One prime materials platform has been the rare-earth nickelates; indeed, superconductivity was recently discovered in the doped compound $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ [1]. Undoped NdNiO_2 belongs to a series of layered square-planar nickelates with chemical formula $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$ and is known as the ‘infinite-layer’ ($n = \infty$) nickelate. Here, we report the synthesis of the quintuple-layer ($n = 5$) member of this series, $\text{Nd}_6\text{Ni}_5\text{O}_{12}$, in which optimal cuprate-like electron filling ($d^{8.8}$) is achieved without chemical doping [2]. We observe a superconducting transition beginning at ~ 13 K. Electronic structure calculations, in tandem with magnetoresistive and spectroscopic measurements, suggest that $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ interpolates between cuprate-like and infinite-layer nickelate-like behaviour. By engineering a distinct superconducting nickelate, we identify the square-planar nickelates as a new family of superconductors that can be tuned via both doping and dimensionality. In this poster, we focus on the superconducting characteristics and electronic structure of the $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ compound and greater family of layered nickelates [3].

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Rare-Earth Nickelates: from Perovskite to Infinite-Layer

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Perovskite rare-earth nickelates ($RNiO_3$) have been investigated in detail because of their rich phase diagram comprising a sharp metal-to-insulator transition, an unusual antiferromagnetic ground state, and the possibility to tailor their properties via heteroepitaxy. Recently, the observation of superconductivity in the Sr-doped infinite-layer nickelate $Nd_{0.8}Sr_{0.2}NiO_2$ by Li et al.[1] intensified the interest in this material class. Subsequently, superconductivity was also found in several related compounds and in the quintuple-layer square-planar nickelate $Nd_6Ni_5O_{12}$ [2]. However synthesis is very challenging. It requires the growth of a doped perovskite precursor or the Ruddlesden-Popper phase with the right stacking and a subsequent reduction to the infinite-layer phase by a soft chemistry topotactic reaction with CaH_2 . The reduction is crucial, but at the same time not well understood. Comparing, for example, Lee et al.'s[3] and Zeng et al.'s[4] papers the listed temperatures and times vary. This contribution will have two main focusses: the growth of perovskite rare-earth nickelates undoped and doped by atomic-layer-by-layer oxide molecular beam epitaxy (MBE) and an investigation of the reduction process. On one hand, using MBE for synthesis is interesting due to the possibility for accurate control, high sample quality and sharp interfaces in heterostructures. On the other hand, perovskite nickelates are difficult to synthesize due to a relatively high Ni oxidation state 3+. Their synthesis requires high pressures, which are limited by the working principle of MBE. Additionally, off-stoichiometry easily results in the growth of different phases[5]. After discussing these challenges, that are further increased by doping, some questions on the reduction process will be addressed through studies of differently far reduced samples. Since superconductivity was only observed in epitaxial thin layers, the question arises to what extent the reduction process is sample-specific here. In particular, lattice defects resulting from the lattice mismatch with the substrate, spatially varying cation stoichiometry as a result of the growth process, and islands resulting from mixed layer-by-layer and island growth typical of these nickelate layers, are areas where an inhomogeneous reduction can occur. Therefore, spatially resolved information about the oxidation state and the oxygen de-intercalation pathways in these compounds may reveal strategies for enhancing superconductivity in the nickelates. To this end a study with a soft x-ray microscope showing lateral homogeneity down to length scales of 50nm[6] will be presented. Furthermore, measurements on effects of stepwise oxygen reduction and doping on the electronic structure will be presented.

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Sharpening the Superconducting Transition of $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ Films with $n=1-3$ grown by MBE

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In bulk, the $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ (i.e., Bi-cuprate) family provides the highest superconducting transition temperature—up to 105 K [1]—without highly toxic constituents like thallium or mercury. When prepared as epitaxial films, however, the superconducting transitions of this family are broad and depressed for as-grown epitaxial films, with the highest zero-resistance T_c reported to date of 97 K [2]. In this work, we use ozone-assisted MBE to grow $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4}$ ($n=1-3$) single-layers and bi-layers on (100) SrTiO_3 substrates, where we use a non-superconducting $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ (Bi-2201, $n=1$) buffer layer to reduce disorder caused by the underlying substrate. We demonstrate that combining adsorption-controlled *co-shuttered* growth and the initial precise flux calibration provides the best structural and superconducting properties for Bi-cuprate films formed at growth rates of 0.1 $\mu\text{m/hr}$ or higher.

In-situ reflection high-energy electron diffraction exhibits incommensurate structural modulation and also proves the absence of any secondary phases. The structural quality of the samples is confirmed using a combination of four-circle x-ray diffraction, atomic force microscopy, and transmission electron microscopy. The superconducting properties are studied by resistivity vs temperature measurements. Bi-2201/Bi-2212 bilayers exhibit the smoothest reported surfaces with subnanometer root-mean-square (rms) roughness of ~ 0.4 nm and the sharpest superconducting transition width (ΔT_c) ~ 10 K, similar to Bi-2212 single crystals. While the $T_{c(R=0)}$ for the as-grown bilayer is low, i.e., ~ 50 K, this is a matter of oxygen content and can be modified via a post-growth process. This presentation focuses on achieving the high-structural and surficial quality and the sharp ΔT_c of the as-grown samples. We conclude that combining the *co-shuttered* growth and the initial precise flux calibration provides the best structural and superconducting properties [3].

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Characterization of the Superfluid Density in Infinite-Layer Nickelates

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Infinite-layer nickelates present a new family of potential unconventional superconductors [1-4]. A key open question is the superconducting gap symmetry. In this presentation, I will present our low-temperature characterization of the superfluid density in $(R,\text{Sr})\text{NiO}_2$ ($R = \text{La, Pr, Nd}$) utilizing the mutual inductance measurement technique [5]. For La and Pr-nickelates, the superfluid density shows a quadratic temperature dependence, indicating nodal superconducting gap structure in the presence of disorder. Nd-nickelate instead exhibits complex low-temperature behavior due to the magnetic contribution of the Nd^{3+} $4f$ moments [6]. These results are consistent with d -wave pairing.

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Rare-Earth Control of the Superconducting Upper Critical Field in Infinite-Layer Nickelates

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The role and consequences of varying the rare-earth element in the superconducting infinite-layer nickelates have been much debated [1-5]. In this work, I will present striking differences in the magnitude and anisotropy of the superconducting upper critical field across the La-, Pr-, and Nd-nickelates. We find that these distinctions originate from the $4f$ electron characteristics of the rare-earth ions in the lattice: they are absent for La^{3+} , nonmagnetic for the Pr^{3+} singlet ground state, and magnetic for the Nd^{3+} Kramer's doublet. We also find unique polar and azimuthal angle-dependent magnetoresistance in the Nd-nickelates, which can be understood to arise from the magnetic contribution of the Nd^{3+} $4f$ moments [6]. In the absence of rare-earth effects, we find that the nickelates broadly violate the conventional Pauli limit. Such robust and tunable superconductivity suggests potential in future high-field applications.

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Physical and electronic structure of square-planar Nickelates

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Square-planar nickelates are a novel class of superconductors, which promise to help unveil aspects about the nature of superconductivity in oxide materials. We characterize the atomic and electronic structure of this class of square planar nickelates grown using molecular beam epitaxy (MBE). Atomic structure is characterized using crystal truncation rod (CTR), and electronic structure is characterized using diffraction-based X-ray absorption near edge structure (dXANES). Both methods of characterization are performed at the same time at beamline 33ID using tunable synchrotron radiation. At different stages in the reduction process, CTR measurements are consistent with a phase transition from oxygen-deficient NdNiO_{3-x} to NdNiO_2 as indicated by a monotonic change in lattice constant from 3.75\AA to 3.30\AA . Changes in the electronic structure measured by dXANES at the Ni K edge shows a monotonic 5eV shift of the white line energy as NdNiO_3 is reduced to NdNiO_2 . We also observe a pre-peak feature 9eV below the white line maximum for NdNiO_2 , which is also observed for the square-planar cuprates.

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The effect of spin-polarized EuTiO₃ on the monolayer FeSe superconductor

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The physics of high-temperature superconductors is governed by the interplay of the spin and charge degrees of freedom. The spin degree of freedom plays an important role in the unconventional superconductivity of cuprates, where a long-range antiferromagnetic phase always exists next to the superconductivity dome. FeSe has the simplest chemical composition among Fe-based superconductors, yet monolayer FeSe on SrTiO₃ substrate reports one of the highest superconducting transition temperatures. Bulk FeSe undergoes a nematic transition with spin fluctuations suggested as the pairing mechanism, but no static magnetic ordering has been observed [1]. Monolayer FeSe grown on oxide substrates exhibits neither nematicity nor magnetic ordering [2].

We grow monolayer FeSe on antiferromagnetic EuTiO₃(001) layers [3, 4], to introduce a spin polarization in proximity to the superconductivity of FeSe. By X-ray absorption spectroscopy, we observe an antiferromagnet-ferromagnet switching in EuTiO₃ driven by an applied magnetic field on both Eu and Ti sites, with no concomitant magnetic ordering induced on the Fe site of FeSe. Transport measurements show the superconductivity of monolayer FeSe survives on EuTiO₃ with a transition temperature of ~ 30 K and an upper critical field of 40 T. The band structures revealed by photoemission spectroscopy are analogous to those of FeSe/SrTiO₃. Our work creates a platform to study the interplay of spin and superconductivity in the two-dimensional limit that aims to provide insights into the unconventional superconductivity of FeSe.

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SrNbO₃ and SrTaO₃ Heterostructures Grown Using Hybrid Molecular Beam Epitaxy

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SrNbO₃ (SNO) and SrTaO₃ (STaO) contain a single d-band electron at the Fermi energy level that is readily transferred across the interface to other materials, such as BaSnO₃ (BSO). It has been theoretically predicted that SNO and STaO would be effective interfacial donor candidates to STO, with Nb donating 0.36 electrons and Ta donating 0.63 per unit cell [1]. To date, however, work developing SNO and STaO has been extremely limited. Here we report our progress on the development of these two materials using hMBE, along with the development of SHO to serve as a dielectric barrier and stable capping layer for atmospheric exposure [2]. Films were characterized using *in vacuo* photoelectron spectroscopy (XPS) was used to characterize the stability of the Nb⁴⁺ and Ta⁴⁺ charge states. *Ex situ* X-ray absorption near edge spectroscopy (XANES) illustrates the importance of the SHO capping layer in preserving the Nb 4d¹ (Nb 4+) metastable charge state for SNO even in atmospheric conditions. XPS valence band data for the Nb 4d electronic states near the Fermi level shows the feature near fermi level indicating electron donor ability of Nb. For STaO, we found semiconducting LaFeO₃ can act as a capping layer for the Ta 5d¹ (Ta⁴⁺) state.

Heterostructures of SNO were also prepared on high mobility semiconductor BaSnO₃ (BSO) thin film surfaces to examine charge transfer between SNO and BSO across the interface. Using a combination of density functional theory (DFT) modeling of the BSO/SNO interface and *in vacuo* XPS, we find sheet carrier densities $\sim 10^{14}$ e-/cm², which are the largest observed for SNO heterostructures. Angle-resolved XPS measurements are in agreement with DFT models and show that the interfacial BSO layer receives ~ 0.3 e- per Sn ion and that there is significant electron diffusion farther into the BSO layer due to the low effective mass and highly dispersive bands of BSO. These results demonstrate that SNO can serve as a strong donor material for future 2DEG structures.

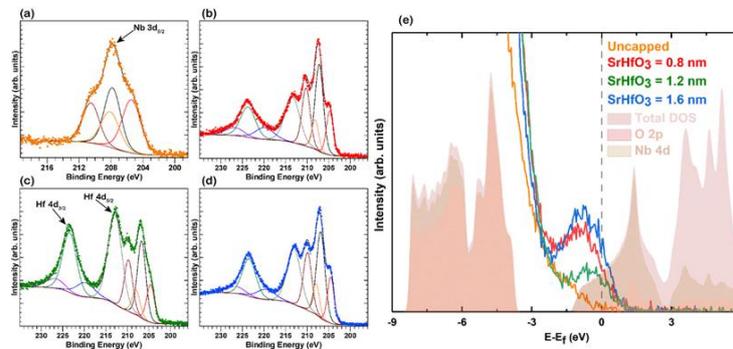


Fig 1: Nb 3d core level deconvolution for SNO thin films capped with (a) uncapped, (b) 0.8 nm, (c) 1.2 nm, and (d) 1.6 nm capping of SHO. Fits to the data show spin-orbit split peaks of Nb⁴⁺ (low binding energy) and Nb⁵⁺ (high binding energy) features. (e) Valence band XPS data showing density of states near the Fermi level for all four samples.

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Identifying a MBE growth window for quaternary melilite oxide films by integrating experimental and computational approaches

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The advancement of the electronic industry is powered by new materials and new physics. In this work, we perform thermodynamic modeling using the CALculation of PHase Diagrams (CALPHAD) approach aided by density functional theory (DFT) calculations to guide the synthesis of $\text{Sr}_2\text{MnGe}_2\text{O}_7$, a quaternary oxide with the melilite structure. We use the modeling results to obtain a growth window for film synthesis by molecular beam epitaxy (MBE). This material has recently been proposed to be a Weyl semimetal [1], and has been previously synthesized in powder form [2]. However, we are unaware of any previous reports of epitaxial melilites heterostructures.

As the final purpose is to obtain accurate thermodynamic information, DFT results obtained using different exchange-correlation functionals were compared in terms of the formation enthalpy of the oxide phases in the Sr-Mn-Ge-O system that are available in the SSUB6 database [3]. The results show that the GGA/GGA+U mixing approach employed in the Materials Project (MP) database [4,5] is the most accurate in predicting the enthalpy of oxidation reactions. Therefore, this approach is employed to estimate the formation enthalpy of the Sr-Mn-Ge-O oxide phases that are not available in the SSUB6 database. A preliminary thermodynamic database of the Sr-Mn-Ge-O system is built from the SSUB6 database for the phases available and the unavailable phases are added based on the formation enthalpies from the MP database, as well as formation entropies from a linear approximation based on the ratio of anions/cations. The results show an adsorption-controlled growth window for $\text{Sr}_2\text{MnGe}_2\text{O}_7$ in which an extra supply of Ge would be desorbed from the surface as a gas species (i.e., GeO). Initial results of MBE synthesis will be presented, and the obtained phases present in the films will be compared against the calculated thermodynamic growth windows.

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Epitaxial growth of the candidate ferroelectric Rashba material SrBiO₃ by pulsed laser deposition

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Among oxides, bismuthates have been gaining a lot of interest due to their unique properties. Historically known for their superconducting behavior, they now show prospects for applications in a wide range of domains, mainly owing to the presence of Bismuth as a source of spin-orbit coupling [1]. The ongoing investigation of bismuthate thin films has so far been centered on BaBiO₃, with studies focusing on the growth of relaxed epitaxial thin-films [2]. Here, we demonstrate for the first time the growth of epitaxial films of SrBiO₃ on both TiO₂-terminated SrTiO₃ ($a=3.905$ Å) and MO-terminated MScO₃ substrates (M=Dy, Nd) by pulsed laser deposition. SrBiO₃ has a pseudocubic lattice constant 4.25 Å and quickly relaxes as thickness increases. High-resolution transmission electron microscopy (TEM) reveals two types of substrate-film interfaces can arise depending on the lattice mismatch. In one case, the formation of mismatch dislocations leads to macro domains with portions of the films subjected to tensile strain. In the other, smoother relaxation occurs over a thicker film of SrBiO₃, which results in partly tetragonal SrBiO₃ films subjected to compressive strain. Our findings regarding the polarization appearing in both type of films are encouraging towards the development of non centro-symmetric SrBiO₃ as a ferroelectric Rashba Semiconductor [3].

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Direct ARPES and STM investigation of various oxide thin films/heterostructures by Pulsed Laser Deposition at APE-NFFA Beamline

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Functional oxide perovskite materials exhibit wide range of phenomenon such as 2-dimensional electron gas at the interface of polar/non-polar heterostructure in LaAlO₃/SrTiO₃, superconductivity, ferroelectricity, ferromagnetism etc. Emergent physical observations can be made in oxide materials when grown in ultra-thin limit with epitaxial strain induced by the substrate or by the interface in heterostructures with other functional materials. Therefore, fabricating oxide thin films with atomically sharp interfaces and flat surfaces is an utmost importance for probing the surface sensitive properties by angle-resolved photoemission spectroscopy (ARPES).

Here, by choosing Pulsed Laser Deposition (PLD) technique equipped with two different lasers namely KrF Excimer laser of 248 nm (UV) and ND:YAG solid-state laser of 1064 nm (IR region) [1], we have successfully demonstrated the controlled growth process of various atomically flat oxide thin film heterostructures. Thanks to the unique facility i.e., the PLD connected in-situ UHV with the ARPES at the NFFA - APE beamline at Elettra Synchrotron, gave us an opportunity to probe electronic states of different oxide surfaces. To name a few, an observation of 2-DEG on the 1 u.c. on the anatase TiO₂ surface [3], a decades old challenges in the understanding of the origin of Metal-Insulator transition in vanadium oxide films which is often wrongly attributed with the structural changes has also addressed.

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Shutter-method growth of (001), (110) and (111)–oriented perovskite films

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The alternating-shutter approach has been shown to be an effective method for the growth of high-quality films of (001) orientation for a wide number of perovskite oxide phases. Compared to codeposition, the shutter method has the advantage of providing precise control of the film stoichiometry and surface termination, *in situ* and in real time, via interpretation of the reflection high-energy electron diffraction (RHEED) oscillations. Due to the increasing interest in the novel properties of interfaces between two perovskites, as well as exploring the properties of monolayer films and membranes, different bonding environments beyond the (001) orientation are being studied, for example the layered hexagonal network of (111) surface. Here we report an extension of the model for interpreting RHEED oscillations originally developed for (001) growth to both (111) and (110) growth directions. We show that the universal model developed for (001) growth via RHEED oscillations can also be extended to control (111) and (110) growth. We present results of shutter growth of nonpolar (e.g. SrTiO₃) and polar (e.g. LaFeO₃) films on substrates of all three orientations, and draw conclusions on the overall utility of the shutter method in these cases.

hMBE growth of BaTiO₃ thin films for electrical and optical applications

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BaTiO₃ is an appealing class of ferroelectric perovskite materials that has a room-temperature ferroelectricity with a Curie temperature, T_c , of ~ 120 °C. It has been extensively studied due to its superior dielectric, ferroelectric, piezoelectric, pyroelectric, and electro-optical properties. Therefore, it has emerged as an excellent candidate for applications such as ferroelectric random-access memory, lead-free capacitors, photonic devices, ferroelectric energy conversion, and electro-optical devices.[1] Furthermore, strained BaTiO₃ exhibits both enhanced T_c and increased remanent polarization when compared to bulk BaTiO₃. However, while strain engineering through heteroepitaxial growth has proven to be a powerful strategy for obtaining new functionalities and reaching higher capacities in complex oxides, achieving controlled and uniform stoichiometry in thin films is equally important, as defects can destroy ferroelectric behavior in materials.[2], [3] Although conventional thin film growth techniques and ultrahigh vacuum growth techniques such as pulsed laser deposition and molecular beam epitaxy (MBE) are capable of producing high-quality oxide films, precise control over stoichiometry still remains challenging. Therefore, *h*MBE which has been demonstrated to provide superior control over cation stoichiometry was used in this study to grow BaTiO₃ thin films.

*h*MBE growth of BaTiO₃ thin films was performed using a solid elemental Ba source and a metal-organic precursor (Titanium(IV) isopropoxide) that was supplied via a gas inlet system. A series of thin films were grown on LSAT ((LaAlO₃)_{0.3}(Sr₂AlTaO₆)_{0.7}), GdScO₃ (110), and SrNbO₃ that was sputtered on GdScO₃ (110) as a transparent bottom electrode due to its stability high temperature and good lattice match with BaTiO₃. In this talk, we will discuss the samples grown within the *h*MBE growth window which yielded films with a constant out-of-plane lattice parameters and atomically smooth surface morphologies in RHEED and AFM. The results of piezoresponse force microscopy and optical measurements such as second harmonic generation and spectroscopic ellipsometry will also be discussed.

This work demonstrates a large avenue to a lead-free ferroelectric for nonvolatile memories and electro-optic devices.

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Machine Learning Analysis of Reflection High Energy Electron Diffraction Images of Epitaxial Oxide Thin Films

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Reflection high energy electron diffraction (RHEED) is a common form of real time analysis used in growth systems such as molecular beam epitaxy (MBE) and pulsed laser deposition (PLD). Traditional RHEED analysis focuses only on the intensity and shape of the diffraction pattern for a few still images taken during growth and is mostly qualitative. While this information has proven insightful, there is far more information that can be gleaned from RHEED. In order to obtain greater insight from RHEED recordings, the novel machine learning techniques principal component analysis (PCA), k -means clustering, and convolutional neural networks were applied to the recordings of the RHEED taken during the molecular beam epitaxy growth of epitaxial thin film perovskite oxides [1,2].

Neural network analysis is extended to predict X-ray photoelectron spectroscopy (XPS) data from RHEED images as a proxy for film composition and material classification. Synthetic XPS data is used to train networks to connect RHEED patterns to XPS data with the eventual goal of estimating film stoichiometry using these neural networks. These methods yield more quantitative results from the RHEED with minimal time requirements and open the door for future development of real-time computer control of film growth for optimal growth conditions.

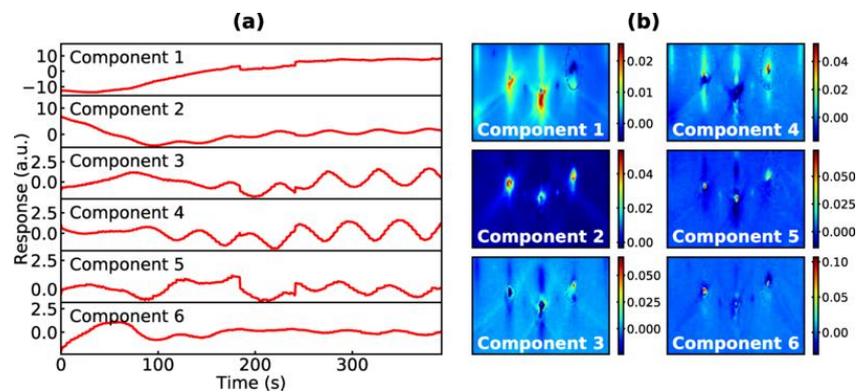


Fig: (a) Principal component eigenvalues as a function of time and (b) the corresponding first six principle component eigenvectors resulting from PCA for a SrTiO₃ film grown on a TbScO₃ substrate.

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***Ex situ* ARPES of $\text{La}_{1-x}\text{Ca}_x\text{NiO}_3$ thin films grown by molecular beam epitaxy**

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The discovery of high- T_C superconductivity in perovskite-based nickelates is driving intense global efforts aimed at explaining the mechanism underlying their unusual properties. “Infinite-layer” doped (112) phase nickelates are structural analogs of the paradigmatic cuprate superconductors and understanding the differences between nickelate and cuprate electronic structures is one of the most important outstanding questions in the field. The topotactic reduction necessary to achieve the (112) phase makes the mapping of the electronic structure on the infinite-layer nickelates by ARPES challenging. However, insight of these materials can be gained by observing the perovskite (113) precursor phase. In the present work, we show the feasibility of obtaining good quality Fermi surfaces and band dispersion in both the doped and undoped LaNiO_3 without the necessity of performing the ARPES study *in situ*. The ability to bring the films in air simplifies their characterization. Buried ARPES of $\text{LaAlO}_3/\text{La}_{1-x}\text{Ca}_x\text{NiO}_3$ heterostructures was also attempted, but resulted in lower quality Fermi surfaces. This project is a collaboration between SBQMI at UBC and the Max-Planck Institute in Stuttgart, Germany.

Hexagallate substrates for oxide spintronic applications

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Modern information and communication technologies will require frequency bandwidths from ~600 MHz up to several terahertz for ultra-fast, low-cost and environmentally friendly data processing. For these challenges, magnetic materials such as single-crystalline barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$) films are attractive for the generation of spin waves used as data carriers in novel computing devices. To achieve epitaxial films with excellent structural and magnetic perfection, a suitable substrate material with small lattice mismatch is required. For the growth of barium hexaferrite by liquid phase epitaxy (LPE), alkaline-earth hexagallates are the substrates of choice, because it has already been demonstrated that they can support the growth of epitaxial films with high crystal quality [1]. If high-quality films could be also grown by molecular-beam epitaxy (MBE), it would bring an opportunity to engineer the properties of hexaferrites using strain engineering. Mateika and Laurien gave the first report on bulk growth of the hexagallate $\text{SrGa}_{12}\text{O}_{19}$ single crystal [2]. It melts peritectically and must be grown from melt solutions with SrO excess. Unfortunately, the growth window from which this phase crystallizes is very narrow; but the authors could increase the yield significantly by an equimolar partial substitution of Ga^{3+} with Mg^{2+} and Zr^{4+} . We were able to show that $\text{Mg}^{2+}/\text{Zr}^{4+}$ co-doping is effective, because it widens the crystallization window [3]. Meanwhile, high-quality bulk crystals of $(\text{Mg,Zr})\text{:SrGa}_{12}\text{O}_{19}$ (SGMZ) with diameters close to one inch (Fig. 1a) were grown at IKZ in the framework of the activities of the Cornell-IKZ joint lab [4] and initial batches of substrates (Fig. 1b) with various sizes were prepared by the company CrysTec GmbH. A novel X-ray diffraction rocking curve imaging procedure specifically developed for SGMZ reveals that the rocking curve widths are typically below 23 arcsec and hence well suited for the preparation of high-quality substrates.

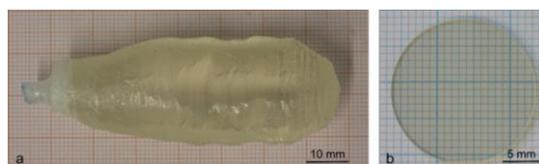


Fig. 1: (a) Typical SGMZ single crystal and (b) prepared substrate with a diameter of 20 mm.

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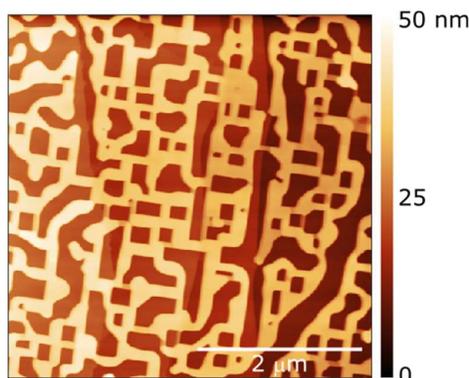
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Dewetting of single crystal oxide membranes

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Recent progresses in oxide growth have substantially advanced the opportunities presented by oxide thin films in both fundamental research and applications. One such technique, which holds significant promise for the future, is the technique to make oxide thin films into freestanding membranes using a variety of etchable buffer layers, inserted epitaxially between the oxide thin film and the substrate during growth. These freestanding membranes can then be transferred to any substrate as long as the membrane adheres to it. Here we transfer SrTiO₃ membranes onto a sapphire substrate using dissolution of a water soluble Sr₃Al₂O₆ buffer layer [1] and subject them to post transfer annealing by heating the substrate to different temperatures. We observe that SrTiO₃ membranes start dewetting from the sapphire at temperatures (T_{dw}) which are significantly lower than the melting point of SrTiO₃. We find T_{dw} depends on the thickness of the membrane, and that the dewetting fronts of the membranes are preferentially oriented along the crystalline axes of the transferred membrane. Furthermore, these do not depend on the relative orientation of the membrane with respect to the sapphire substrate. We also observe that upon increasing the annealing temperature, the dewetting front recedes so far that the film coverage becomes discontinuous, resulting in island formation. During this process, some islands may remain partially connected, resulting in the highly oriented single crystalline nanowires, the formation of which we observe in a repeatable manner on these membranes. These studies can provide a useful platform to study surface energies of different oxides, but also present a useful pathway for micro and nano-patterning [2] of oxides in a non-destructive manner.



An atomic force microscopy image taken, post annealing, of an initially 6 nm thick SrTiO₃ membrane transferred onto a sapphire substrate. The sapphire step terraces can still be seen in the background. The dewetting of the membrane can be seen to have a preference to dewet along a crystalline axis, forming sharp crystalline facets along the receding front of the membrane. This membrane was annealed at a temperature of 1100 °C in presence of 0.075 mbar of oxygen.

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Towards ultraclean correlated metal CaVO_3

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Correlated electron systems have recently come into focus given their potential as novel transparent conductor material for photovoltaics and lighting technologies, but also because some fundamental questions in material systems, in which itinerant carriers experience a sizeable Coulomb interaction, are still far from being understood. New experimental insights require extremely clean material systems to allow directly probing and characterizing the electronic ground state and low energy excitation spectrum of such correlated electron systems. While the correlated metal SrVO_3 with cubic perovskite structure has been grown by hybrid molecular beam epitaxy with a high level of perfection, which resulted in a very high residual resistivity ratio (RRR) in films exceeding values of 200,¹ CaVO_3 films grown by the same growth approach were found to exhibit much lower residual resistivity ratios not exceeding 40. This much lower RRR value of CaVO_3 is not intrinsic in nature, but has been found to originate from a high density of stacking faults formed in CaVO_3 when grown on LaSrAlO_4 substrates, which provide a good in-plane, but poor out-of-plane lattice match.² Stacking faults, misfit dislocations, and twin domain formation of the orthorhombically distorted CaVO_3 can be controlled by growing it epitaxially on a substrate with a more favorable out-of-plane lattice match.

In this talk, we will discuss the growth and transport properties of CaVO_3 thin films deposited on lattice-matched LaAlO_3 substrates using the self-regulated growth mechanism afforded by hybrid molecular beam epitaxy, in which elemental Ca and the V-containing metalorganic precursor vanadium-oxo-isopropoxide (VTIP) were co-supplied to the substrate. A growth window was established, where the out-of-plane lattice parameter of coherently strained CaVO_3 was found to be constant over a wide range of Ca/VTIP flux ratios. Reciprocal space maps confirmed that 55-nm-thick CaVO_3 films grown on LaAlO_3 were coherently strained, and atomically smooth surfaces for the films deposited inside the growth window confirmed the high quality of the series of films grown. Residual resistivity ratio of ~ 100 in CaVO_3 was achieved, indicating excellent structural quality. Correlation of temperature-dependent resistivity and Hall coefficient measurements with the growth conditions and structural properties of the film will be reported. The emergence of a nonlinear Hall effect as a function of temperature and defect density, quantified by the RRR ratio of the films investigated, will be discussed.

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Effect of CMP process on Ba doped PZT deposited by PLD

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In recent years, piezoelectric materials have attracted particular attention for actuation and strain generation in microelectronic systems. Among these materials, lead zirconate titanate **Pb(Zr_{1-x}Ti_x)O₃ (PZT)** has been extensively researched due to its strong piezoelectric response. However, the considerable surface roughness of PZT films renders it difficult to guarantee device reliability, which is one of the major obstacles to the usage of PZT in micromechanical and microelectronic devices.

A proposed strategy to resolve this issue on PZT thin films fabricated by sol-gel technique [1] and MOCVD [2] is to use a **chemical mechanical polishing (CMP)** procedure to planarize the PZT ferroelectric layer. This method involves a chemical dissolution process with a liquid slurry and mechanical abrasive interaction to smoothen and planarize the surface. In this study, 600 nm thick **Ba-substituted PZT [(Ba_{0.1}Pb_{0.9})(Zr_{0.52}Ti_{0.48})O₃] (BPZT)** thin films were deposited by pulsed laser deposition (PLD). PLD has received much interest for its ease of use and success in depositing high-quality materials with complex stoichiometry. These thin films were then polished by a CMP process using a commercial silica slurry; the post-CMP cleaning consisted of a 10-minutes soak in SC-1 solution at 75°C followed by a 5 min soak in IPA in an ultrasonic bath to eliminate slurry residues.

A comparison of BPZT surface roughness (measured by **atomic force microscopy; AFM**) under different CMP settings indicated that the starting root-mean-square (RMS) roughness of **12.14 nm** for 600 nm thick as-deposited BPZT could be reduced to a final roughness below 1 nm (**RMS = 0.79 nm**). These results were lower compared to previous studies using the same slurry [1]. **X-ray photoelectron spectroscopy (XPS)** was used to examine surface composition and contamination after CMP and cleaning. The results show that, after the polishing process, there was a (slurry-related) Si contamination at the surface of the sample, which was removed by the cleaning step. Furthermore, the stoichiometry of Ti, Zr, Pb and Ba did not show a particular dependence on the polishing process.

To confirm that the CMP had no negative impact on BPZT dielectric, ferroelectric, or piezoelectric properties, polarization measurements on BPZT thin films were performed in collaboration with **Solmates (Netherlands)**. Furthermore, the dielectric response of the layer was characterized at microwave frequencies up to 35 GHz by using a basic **concentric capacitor (CC)** architecture. From these measurements, the **dielectric permittivity, ϵ_r** , and the **loss tangent, $\tan\delta$** , for BPZT as deposited and after CMP were determined for different thin film thicknesses. All these measurements reveal that the CMP procedure had little impact on the BPZT characteristics. Moreover, the obtained dielectric permittivity values were higher than in previous studies [3] and decreased less with the BPZT thickness. These findings indicate a reduced impact from the **dead layer** in the stack under investigation, which could be attributable to a better electrode-ferroelectric material interface [4].

F. L. acknowledges support and help by Solmates during the PLD process, and funding by The Research Foundation Flanders (FWO) – grant number 1183722N.

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Epitaxial thin-film realization of an improper ferroelectric spin-liquid candidate

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Magnetically frustrated materials offer a playground for realizing exotic magnetic ground states such as quantum spin ices and spin liquids. Synthesizing such quantum magnets in thin-film form allows further tuning of the magnetic ground state with dimensionality and epitaxial strain as well as the possibility to integrate these states with other functional materials. That said, thin-film quantum spin liquid materials remain scarce. Here, we use reactive oxide molecular beam epitaxy to synthesize the first thin films of hexagonal TbInO₃, a magnetically frustrated rare-earth system that was recently proposed as a spin liquid candidate. TbInO₃ exhibits a geometrically driven improper ferroelectric state, similar to the hexagonal manganites, and the related lattice distortion may play a role in defining its magnetic frustration. Here, we investigate this ferroelectric distortion in epitaxial TbInO₃ thin films using in-situ RHEED and post-deposition HAADF-STEM. We find that the lattice distortion driving the improper polarization appears already at the elevated growth temperature. We furthermore use SQUID magnetometry to investigate the low-temperature magnetic behavior of the Tb³⁺ lattice, where neither long-range order nor spin-freezing is observed, consistent with bulk crystal data. Our work constitutes one of the few thin-film realizations of a quantum spin liquid candidate, and opens up for the use of epitaxy to further manipulate the unusual coexistence of ferroelectricity and spin liquid physics in this thin-film system.

Symmetry Stabilization of High- k Orthoferrite Thin Films

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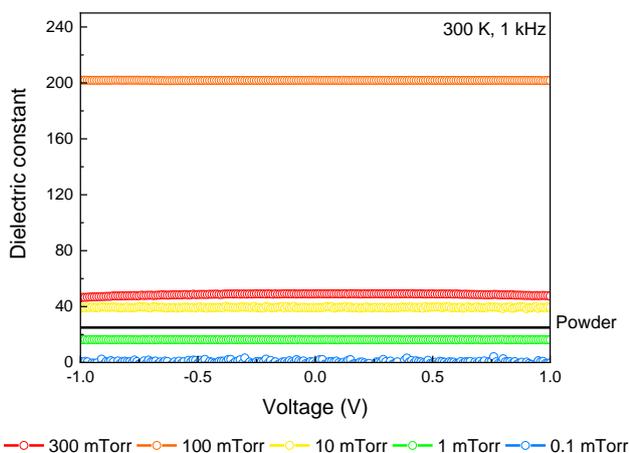
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Strong electromagnetic correlation in insulators can give birth to magnetic high- k dielectric materials essential for oxide electronics and magnetics applications [1]. Orthoferrite perovskite LaFeO₃ is a canted antiferromagnetic insulator at room-temperature, and hence, it serves as a model system for investigating the electromagnetic coupling [2,3]. Most of the studies on LaFeO₃ so far have been limited to bulk polycrystals and powder samples as the research focused mostly on extrinsic effects such as intra, inter-grain effects. Therefore, efforts to observe and control the intrinsic electromagnetic properties of LaFeO₃ in single crystalline form would be highly necessary. Recent advances on pulsed laser epitaxy grants various tunabilities of the functional properties in the thin film growth stage. Especially the structure-property relation in perovskite oxides let us efficiently modify the crystalline symmetry for the functionality tuning [4-6]. Particularly, the control of the crystalline symmetry is expected to derive microscopic magnetic ordering changes and enhancement in the dielectric properties in LaFeO₃ epitaxial thin films.

Here, we achieved epitaxial growth of tetragonal high- k LaFeO₃ thin films with canted antiferromagnetic ordering. We choose the SrTiO₃ (001) substrate with high crystalline symmetry for delivering the symmetry constraint toward the thin films. The tetragonal LaFeO₃ thin films further exhibit modulated electric and magnetic properties by fine control of the stoichiometry. In particular, the dielectric constant of LaFeO₃ thin films was largely enhanced to ~200, from ~25 for conventional bulk powder, as shown in the Figure below [7]. The symmetry control of the LaFeO₃ single crystalline thin film can naturally be extended to more assorted crystalline symmetry control by heterostructuring with other orthoferrites such as SmFeO₃ and ErFeO₃ [8], and interesting modulation of the room-temperature canted antiferromagnetic ordering is expected originating from their structures with controlled orthorhombic distortions.



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Figure 1. Dielectric constants of the epitaxial LaFeO₃ thin films grown at various oxygen partial pressures (P(O₂)). At P(O₂) = 100 mTorr, the largest value of ~200 is achieved, which is 8 times larger than the value for powder sample of ~25 [7].

Tuning the properties of Cathodic Vacuum Arc Deposited Vanadium Oxides thin films

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Among transition metal oxides (TMO), vanadium-based oxides are found in many Magneil and Wadsley phases showing interesting properties. In this study, for the first time we report V_7O_{16} phase of vanadium oxide thin films grown by post-annealing of cathodic vacuum arc- deposited amorphous vanadium oxide thin film on barium-borosilicate glass (7059). A permanent structural phase transformation from V_7O_{16} to V_2O_5 was seen around ~ 400 °C annealing temperature [Figure 1]. The structural, electronic and optical properties of these films were investigated using X-ray Diffraction, Raman Spectroscopy, Resistance vs. Temperature measurement, UV-Visible, Photoluminescence (PL) and Time Resolved Photoluminescence (TRPL) spectroscopies. The conductivity, transmission and the colours of these films could be tuned significantly by sweeping the temperature. We explain these results through the static and dynamic structural changes and the role of oxygen vacancies. These results will be presented and discussed.

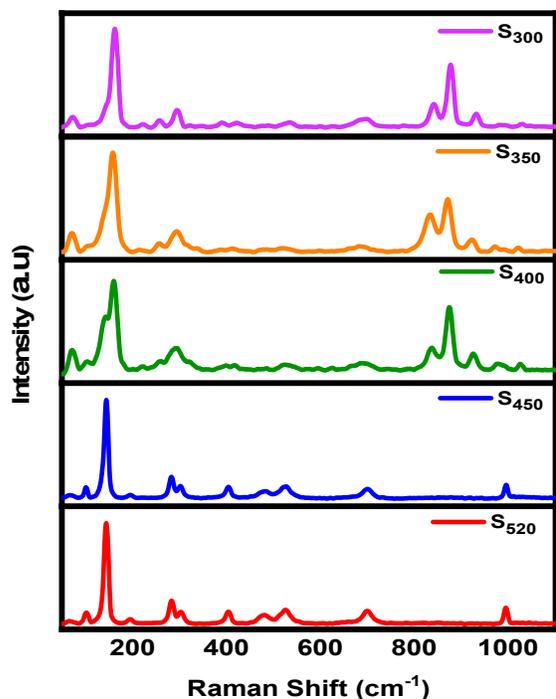


Figure 1: Raman Spectra of Vanadium Oxides annealed at various temperature.

Growth of Potassium Tantalate Films by Suboxide-MBE

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Complex transition metal oxides exhibit an incredible variety of orders and instabilities with a wide range of technologically-relevant physical phenomena including colossal magnetoresistance, high temperature superconductivity and highly tunable magnetism and ferroelectricity[1–3]. KTaO_3 is an incipient ferroelectric in which a highly anisotropic superconductivity emerges near a polar instability in electron doped samples. KTaO_3 has a very large spin-orbit coupling and exotic properties. Emergence of mixed-parity superconductivity and topological Weyl superconductivity are predicted in polar superconductors [4,5]. Growth of high-quality epitaxial films provides an opportunity to control crystal structure and finely tune electronic and polar instabilities.

The use of molecular-beam epitaxy to grow KTaO_3 has evaded demonstration—until now. Using a molecular beam of the suboxide TaO_2 emanating from an effusion cell containing Ta_2O_5 in combination with a molecular beam of potassium emanating from an indium-potassium intermetallic in an oxidant ($\sim 10\% \text{O}_3 + 90\% \text{O}_2$) background pressure of 1×10^{-6} Torr, KTaO_3 films were grown under conditions of excess potassium. Laue fringes confirm that the films are smooth with a well-defined thickness. Atomic force microscopy reveals atomic steps at the surface of the grown films. Reciprocal space mapping shows the epitaxial KTaO_3 films are coherently strained to the GdScO_3 (110)_o substrate.

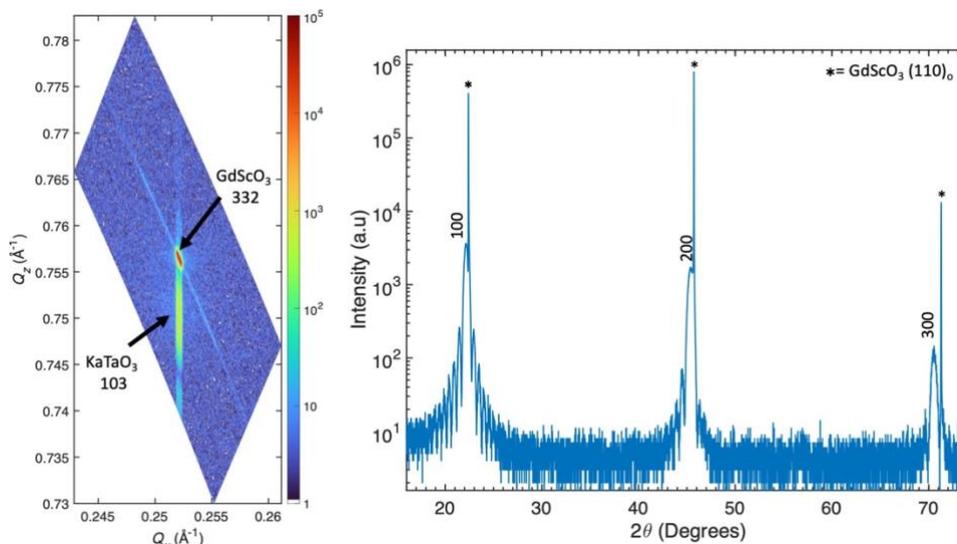


Figure 1: (left) Reciprocal space map of the 103 KTaO_3 peak and the 332 GdScO_3 substrate peak. (right), θ - 2θ x-ray diffraction scan of the same 17 nm thick KTaO_3 film grown on GdScO_3 (110)_o.

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Opening for Device Integration of RENiO_3 Thin Films by Low Temperature Direct Epitaxy by Atomic Layer Deposition

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Complex oxide electronics are attracting increasing attention due to the possibility of constructing devices with fundamentally different behavior, as compared to their traditional silicon counterparts. A crucial challenge for the implementation of complex oxide electronics has been the lack of applicable deposition techniques that work under conditions that can facilitate monolithic device integration, and on challenging substrate geometries.

High quality epitaxial complex oxides are typically deposited by physical techniques such as pulsed laser deposition or molecular beam epitaxy. While these techniques offer extremely high quality films and sharp interfaces in a lab setting, they typically operate at high temperatures and vacuum, and on limited substrate areas. Chemical vapor deposition (CVD) is also employed, but the resulting films are typically in need of post deposition annealing at 600+ °C to facilitate solid phase epitaxy.

This has traditionally also been the case for the CVD daughter technique atomic layer deposition (ALD), but with new insight in growth we have been able to find routes for direct epitaxy of a range of nickelates and ferrites at temperatures as low as 225 °C. ALD offers straightforward upscaling on large area wafers with already existing deposition technology. Furthermore, the technique does not require line of sight, and can be used to deposit films on substrates with complex geometries.

In this talk, we present a route for direct epitaxy of exotic rare earth nickelates at temperatures as low as 225 °C. The resulting thin films, exemplified by LaNiO_3 , NdNiO_3 , EuNiO_3 and GdNiO_3 , are highly epitaxial as deposited on LaAlO_3 (100/110/111) and SrTiO_3 (100/110/111), with no post-deposition annealing required to obtain the attractive electronic properties. The interface towards the substrates are near defect free, and extremely sharp. Thickness can be varied between sub-nanometer and approximately 100 nm without loss of epitaxy or functionality. For LaNiO_3 , Hall measurements reveal room temperature resistivities in the 100 $\mu\Omega$ cm range, with carrier concentrations $>10^{22}$ cm^{-3} . For NdNiO_3 , a sharp metal-insulator transition (MIT) is found at 140 °C, spanning two orders of magnitude for their resistivity. GdNiO_3 is found to be metastable, and the low deposition temperature is crucial to avoid decomposition.

We go on to show how ALD can further be used to cation substitute the host nickelates on either A- or B-site to tune the electronic properties. This is exemplified by B-site Cu-substitution in LaNiO_3 and A-site Sm-substitution in NdNiO_3 . In $\text{LaCu}_x\text{Ni}_{1-x}\text{O}_{3-x}$, we show that electronic resistivity can be tuned smoothly over *six* orders of magnitude from 100 $\mu\Omega$ cm to 100 Ω cm, concomitantly showing increasing MIT temperatures tunable from 0 K to above RT. In NdNiO_3 , Sm-substitution can be used to increase MIT towards room temperature.

This new development in the possibility of low-temperature direct epitaxy of important functional materials open up for facile monolithic device integration of complex oxides, which will become important in novel oxide electronic devices.

Physical properties of *n*-type EuTiO₃ thin films with La³⁺ (4*f*⁰) and Gd³⁺ (4*f*⁷) donors grown by gas source molecular beam epitaxy

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EuTiO₃ (ETO) is a unique magnetic semiconductor with a large magnetic moment of Eu²⁺ (4*f*⁷) where electrons can be easily doped in Ti 3*d* conduction band by chemical substitution. Recently, the development of a gas source molecular beam epitaxy technique enabled to fabricate exceedingly high mobility ETO films hosting peculiar magnetotransport properties such as unconventional behaviors in anomalous Hall effect originating from topological band structure as well as Shubnikov-de Haas oscillations in forced ferromagnetic state [1, 2]. So far, magnetotransport properties have been investigated for electron doped ETO by chemical substitution of several tri-valent rare earth ions such as La³⁺, Sm³⁺, and Gd³⁺ for Eu²⁺ site. For example, when non-magnetic donor La³⁺ is doped to ETO, the spin-less site is introduced to the spin lattice composed of Eu²⁺ magnetic moment. Nevertheless, the influence of spin defects on physical property such as spin scattering had not been investigated systematically in previous studies. Here, we have studied the physical properties of ETO films with La³⁺ (4*f*⁰) and Gd³⁺ (4*f*⁷) donors to investigate the influence of spin defects at Eu-site.

Epitaxial thin films of doped ETO, namely Eu_{1-x}Gd_xTiO₃ (EGTO) and Eu_{1-x}La_xTiO₃ (ELTO), were grown on (001) SrTiO₃ (STO) substrates with a gas source molecular beam epitaxy. As for magnetic properties, the Néel temperature (*T*_N) of EGTO keeps almost constant against the Gd doping, while that of ELTO is reduced with increasing the doping concentration as seen in Fig. 1 (a), presumably due to the vacancy of magnetic moment on La site. As shown in Fig. 1 (b), the maximum electron mobility is larger for ELTO (3,200 cm²V⁻¹sec⁻¹) than EGTO (1,500 cm²V⁻¹sec⁻¹) with keeping metallic state at very diluted doping range, indicating local buckling of TiO₆ octahedron for smaller ion of Gd³⁺ doping case gives stronger scattering. In this presentation, the magnetic field dependence of transport and magnetization will be also discussed in detail.

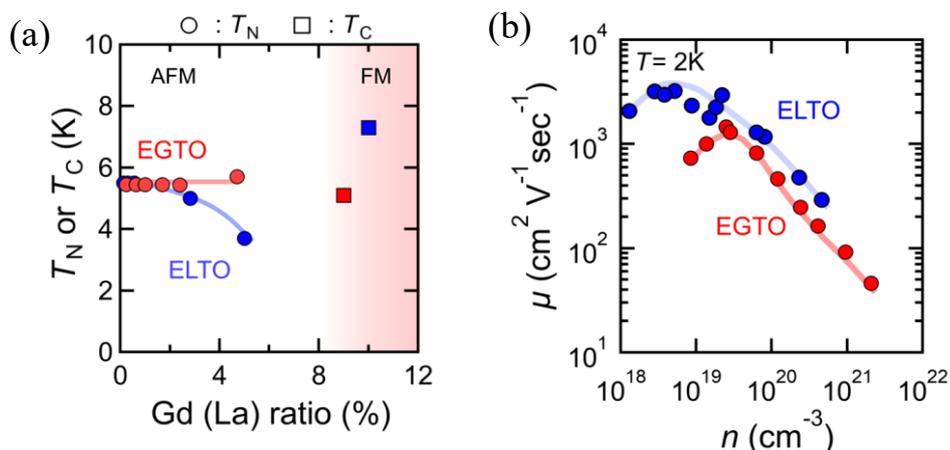


Fig.1 (a) Néel (*T*_N, circle symbols) and Currie (*T*_C, square symbols) temperatures for EGTO (red) and ELTO (blue) films. (b) Carrier density dependence of mobility (*μ*) for EGTO (red circles) and ELTO films (blue circles).

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Growth and study of $\text{CaCuO}_2/\text{SrTiO}_3$ bilayer thin films

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The observation of high T_c superconductivity in doped infinite-layer cuprates (ACuO_2 with $A=\text{Ca}$, Sr or Ba) [1] has been subject of intense research since its discovery.

Recently, interface superconductivity was observed in $\text{CaCuO}_2/\text{SrTiO}_3$ bilayers [2]. It was shown that the doping leading to superconductivity originates from the presence of apical oxygen atoms located in the calcium planes at the interface. The insertion of apical oxygen is related to the proximity of the SrTiO_3 thin film whose perovskite structure promotes additional oxygen in the infinite-layer during the growth [3].

In this work we use pulsed laser deposition to grow $\text{CaCuO}_2/\text{SrTiO}_3$ bilayer thin films. We explore various growth parameters (pressure, temperature, O_2/O_3 ratio, gas flow...) with the aim to obtain superconducting interfaces. The structural and electronic properties are investigated by X-ray diffraction and electrical transport measurements.

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***In situ* soft chemistry engineering of nickelate superconductors**

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Superconducting infinite-layer nickelate thin films are obtained by topotactic reduction from the perovskite precursor phase [1]. In aim of improving the understanding and control of the oxygen deintercalation reaction, and improving the quality of superconducting nickelate thin films, we have developed an experimental platform that can monitor the topotactic reduction by transport measurements between 620 K to 1.7 K. Precise control of the chemical reaction provides a new way to tune the electronic properties of nickelate superconductors. The technique of *in situ* soft chemistry engineering also opens the possibility of exploring emergent quantum phases in other oxide thin films that can undergo topochemical reactions.

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Investigation of Sn^{2+} compounds as candidate p -type conducting oxides

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Over the past several decades, engineering of n -type semiconducting oxides has advanced steadily from material discovery to device engineering while the complementary p -type oxides appear perpetually stuck in the material discovery phase of development. In this work, we use molecular-beam epitaxy (MBE) to synthesize epitaxial thin films of an emerging candidate p -type oxide, Ta_2SnO_6 , for the first time [1]. A central challenge in epitaxially synthesizing Ta_2SnO_6 phase was selecting a substrate to seed growth of the monoclinic crystal structure (Fig. 1 (a)). Ultimately, adsorption-controlled synthesis of high quality Ta_2SnO_6 films was demonstrated on (101)-oriented TiO_2 substrates (Fig 1(d)), leveraging the observation that Ta_2O_5 layers within the Ta_2SnO_6 crystal resemble rutile Fig. 1(a,c) [2]. This synthetic achievement enabled detailed characterization of this seldom-studied phase. We find that we are unable to induce hole conductivity into these films due to the spontaneous formation of hole-compensating oxygen vacancies under MBE-accessible growth conditions, but calculations indicate that experimental conditions exist for which Ta_2SnO_6 is thermodynamically stable and oxygen vacancies do not form spontaneously [1]. Inspired by the success of our substrate selection, we also use rutile substrates to epitaxially synthesize the first (110)-orientated SnO for the first time (Fig. 1(b)), which self assembles into a forest of epitaxial nanowires.

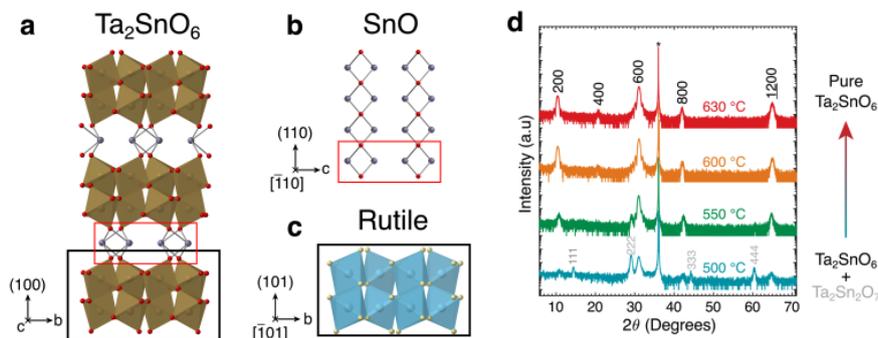


Figure 1 Crystal structure schematics of (a) Ta_2SnO_6 (b) SnO and (c) Rutile (MgF_2). Sn^{2+} , O^{2-} , and F^- ions are indicated by purple, red, and beige spheres. TaO_6 and MgF_6 octahedra are colored brown and blue. (d) XRD θ - 2θ scans of a series of samples grown at varying substrate temperature highlighting the adsorption-controlled growth of Ta_2SnO_6 .

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Nanoscale strain control of the metal-insulator transition in VO₂ thin films

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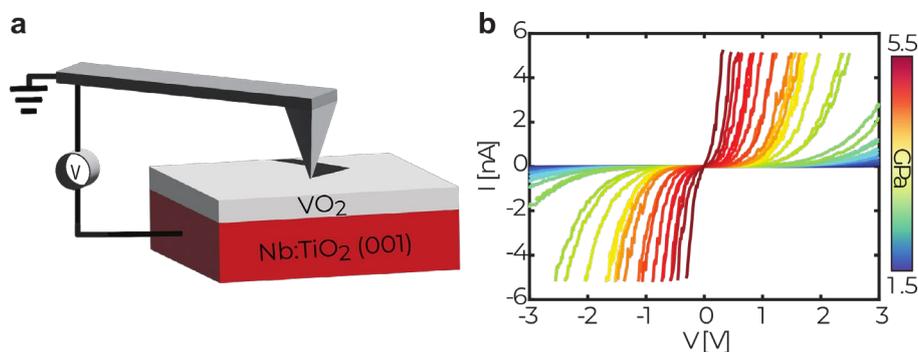
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Nanoscale-induced strain in materials by indenting with a sharp tip can lead to tremendous changes in the local structural and electronic properties. Vanadium dioxide (VO₂) presents an iconic metal-insulator transition (MIT) with a large resistance change near room temperature. The transition can be triggered by various stimuli such as heat, light, and pressure. Uniaxial compression of the rutile c-axis is known to lower the phase transition temperature from the bulk value of 65 °C down to room temperature.

Here, we use conductive atomic force microscopy (C-AFM) to locally modify the electronic properties of a VO₂ thin film by applying specific amounts of pressure to the probe tip. The VO₂ thin film is grown using pulsed laser deposition and epitaxially strained on a semiconducting Nb:TiO₂ (001) substrate. Thanks to the nanometer-sized contact area between the diamond probe tip and the sample, very high pressures of several GPa can locally be applied to compress the lattice of VO₂ in the out-of-plane direction by > 1 % to non-invasively trigger the phase transition at room temperature.

From the obtained current-voltage (I(V)) spectroscopy data, an abrupt transition in the barrier height is observed as a function of the applied pressure and temperature, which we relate to the MIT. We explore the phase coexistence line that separates the insulating and metallic phases in a hitherto inaccessible and unexplored region in the pressure-temperature phase diagram of VO₂. In this work, we restrict ourselves to the low bias and elastic regime, where the transition in transport properties is fully reversible, and we do not observe indications of an electrochemical modification or plastic deformation of the sample.

The combination of epitaxial in-plane confinement with local, uniaxial-like out-of-plane strain due to nanoindentation with a scanning probe tip is a versatile platform for the study of quantum materials under symmetry-breaking strains. The method can readily be extended towards very high electric fields, massive pressure levels, and different ambients.



a Schematic illustration of the nanoindentation of an epitaxially grown VO₂ thin film,

b I(V) spectroscopy as a function of applied pressure between probe tip and sample revealing large changes.

This work is part of the project *Green ICT* (grant number 400.17.607) of the research program NWA which is financed by the Dutch Research Council (NWO).

Anomalous Strain Relaxation and its Impact on the Valence-Driven Spin-State/Metal-Insulator Transition in Epitaxial $(\text{Pr}_{1-y}\text{Y}_y)_{1-x}\text{Ca}_x\text{CoO}_{3-\delta}$ Films

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Pr-based cobaltites such as $\text{Pr}_{1-x}\text{Ca}_x\text{CoO}_{3-\delta}$ and $(\text{Pr}_{1-y}\text{Y}_y)_{1-x}\text{Ca}_x\text{CoO}_{3-\delta}$ exhibit first-order coupled spin-state/metal-insulator/structural transitions driven by a unique Pr valence transition. While restricted to cryogenic temperatures in bulk, our recent work stabilized this valence transition to room temperature (at $T_{\text{vt}} = 291$ K) in $(\text{Pr}_{1-y}\text{Y}_y)_{1-x}\text{Ca}_x\text{CoO}_{3-\delta}$ films grown on YAIO_3 (under 2.1% compressive strain) [1]. Here, we explore strain relaxation in such films. Temperature-dependent resistivity measurements (e.g., Fig 1(a)) reveal surprising splitting of the valence transition into *two* transitions beyond a critical thickness ($t_{\text{crit}} \approx 30$ nm), one at the strained T_{vt} and one near the bulk T_{vt} . X-ray diffraction measurements (e.g., reciprocal space maps (Fig 1(b)) confirm anomalous strain relaxation, where a fully-strained film portion coexists with a near-fully-relaxed portion, *immediately above* t_{crit} . Remarkably, scanning TEM (Fig 1(c)) then reveals a uniformly-spaced dislocation array pinned at the critical thickness, with a fully-strained film beneath. This extraordinary strain relaxation mechanism will be discussed in terms of the unique features of this system, particularly structural and chemical heterogeneity. Similarly surprising findings arise in the ultrathin limit, below ~ 10 unit cells, where T_{vt} remains constant but the resistivity change at the transition is suppressed, destroying the high-temperature metallic state. These results yet further highlight the unique interest and opportunities in these Pr-based valence transition systems.

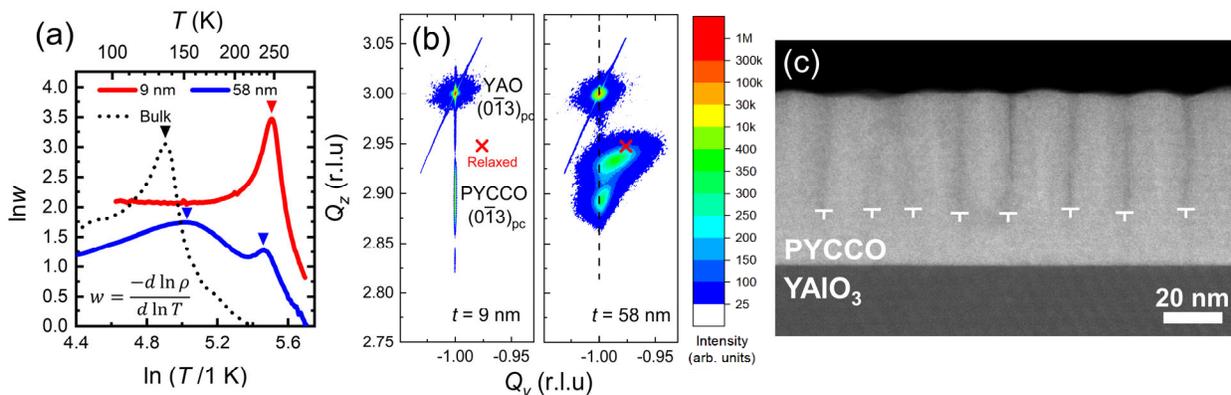


Figure 1: (a) Zabrodskii plot ($w = -d\ln\rho/d\ln T$) of temperature-dependent resistivity (ρ) of $(\text{Pr}_{0.85}\text{Y}_{0.15})_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ films of two thicknesses on YAIO_3 , compared to bulk. Valence transitions are indicated by triangles. (b) Asymmetric X-ray reciprocal space maps illustrating fully-strained growth of $(\text{Pr}_{0.85}\text{Y}_{0.15})_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ on YAIO_3 (left), and division into fully-strained and near-fully-relaxed portions above t_{crit} (right). (c) Cross-sectional scanning TEM of the 58-nm-thick $(\text{Pr}_{0.85}\text{Y}_{0.15})_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ film in (a) and (b), with periodic edge dislocation cores labeled.

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Disentangling the insulator-to-metal and structural transitions in VO₂ thin films with non-contact atomic force microscopy

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Vanadium dioxide (VO₂) undergoes a thermally driven insulator-to-metal transition with a resistivity decrease up to five orders of magnitude when heated above 341 K, making it a promising material for oxide electronic applications. This transition is associated with a change in crystal structure from monoclinic symmetry below the transition to a rutile structure above the transition. Despite more than 60 years of research, whether the insulator-to-metal transition originates from the structural change that doubles the unit cell (i.e., Peierls distortion) or by electron correlation effects that push the system to become insulating (i.e., Mott transition) remains an open question. In addition, an insulator-to-metal transition has also been observed in voltage-biased thin films as a sharp jump in the measured current response when the voltage is increased. The interplay between Joule heating and electric field effects in inducing the insulator-to-metal transition is an active area of research.[1]

In this work, we use oxygen plasma-assisted molecular beam epitaxy to deposit 10 nm thick films of VO₂ on (001) TiO₂ substrates. *Ex-situ* x-ray diffraction, atomic force microscopy, and electrical transport measurements confirm the films are epitaxial with atomically smooth surfaces and a large resistivity change at the insulator-to-metal transition. In our films, the insulator-to-metal transition temperature is reduced to around 290 K due to substrate-induced strain, in agreement with previous reports.[2]

To directly probe the insulator-to-metal transition, we performed *in-situ* non-contact atomic force microscopy (nc-AFM) measurements on the VO₂ films. In our nc-AFM, we use an oscillating qPlus sensor [3] to locally detect the dissipative forces between the tip and the sample. To compensate for these dissipative forces, which act to dampen the oscillation amplitude of the tip, we continuously drive the system to maintain a constant oscillation amplitude. By quantitatively tracking the power required to maintain the oscillation amplitude as a function of sample temperature, the voltage applied between the tip and the sample, and the tip-sample distance, we can delineate several regimes with distinct dissipation signatures, which correspond to different dominant tip-sample interactions. By distinguishing between electronic and phononic dissipation channels, our work aims to disentangle the coupled electronic and structural transitions in VO₂ thin films.

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Spatial control of the conductivity in SrTiO₃-based heterointerfaces using inkjet printing

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Interfaces between complex oxides host a plethora of functional properties including enhanced ionic conductivity[1], gate-tunable superconductivity[2] and exotic magnetic states[3]. The enhanced electronic, ionic and magnetic properties along the oxide interfaces are generally exploited in functional devices by spatial confinement of ions and electrons. Different patterning methods have been used as a way to spatially control the conductivity at the interface.[4,5] One common obstacle for all of these are the multiple steps needed in order to fully fabricate the devices. In this investigation, inkjet printing of thermally stable oxides is introduced as an alternative pathway for spatially controlling the interface conductivity. We inkjet print yttrium-stabilized zirconia and TiO₂ with various shapes and use these as physical masks to confine the electronic conductivity in SrTiO₃-based heterostructures. By performing in-situ measurements of the electrical conductivity as LaAlO₃ and γ -Al₂O₃ are deposited on SrTiO₃, we witness the birth of the interface conductivity and find a consistent transient behavior as conductivity emerges in patterned and non-patterned heterostructures. Interestingly, we find that in both cases conductivity emerges after even the first laser pulse in the pulsed laser deposition in contrast to previous studies. The study paves an alternative way for realizing energy devices with spatially controlled electronic and ionic interface conductivity.

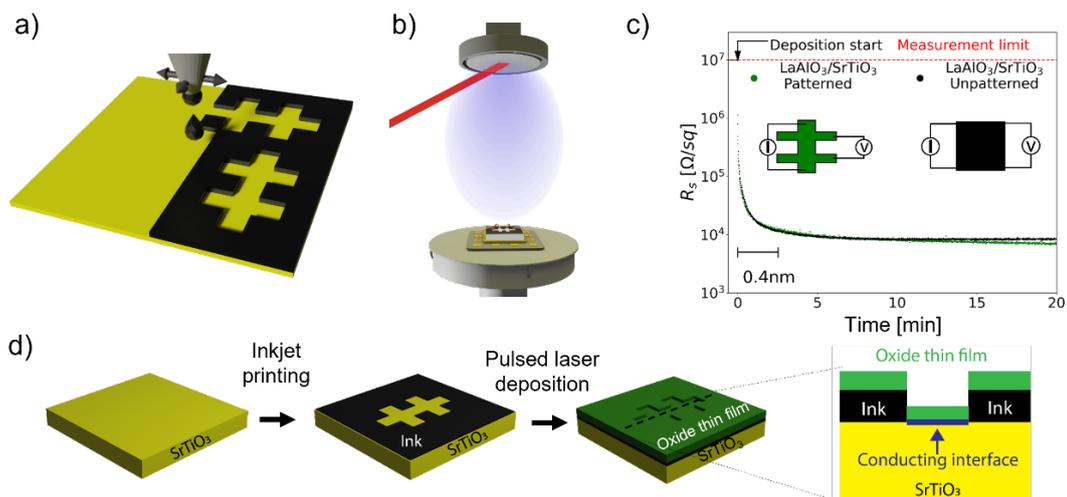


Figure 1: Using inkjet printing to create spatial control of the conducting 2DEG in oxide interfaces. On an empty SrTiO₃ substrate, TiO₂-ink is jetted as a negative mask, a). Subsequently, LaAlO₃ is deposited on top using pulsed laser deposition, b), while in-situ sheet resistance is measured, c). The whole process is depicted in d).

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Electron transport and superconductivity in free-standing LAO/STO micro-membranes on silicon

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Thomas Sand Jespersen¹

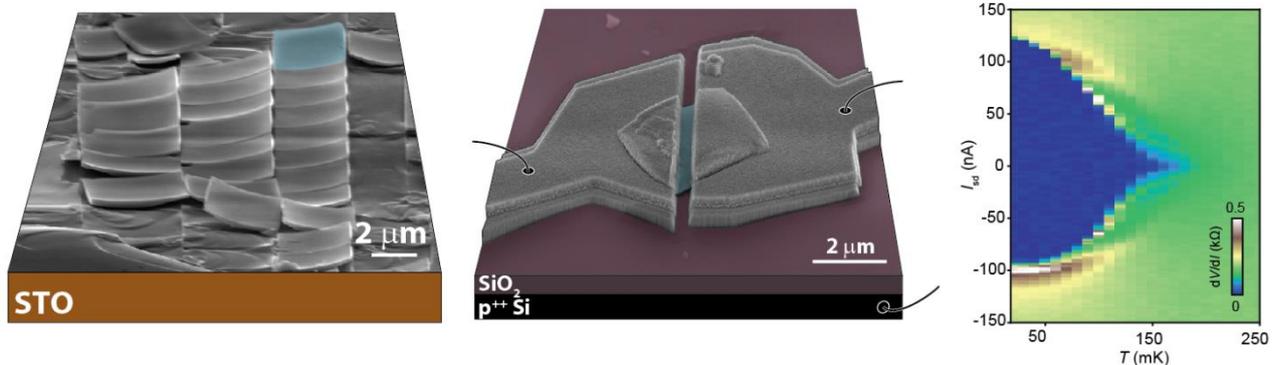
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Freestanding oxide membranes constitute an intriguing material platform for new functionalities and allow integration of oxide electronics with technologically important platforms such as silicon. Sambri et al. recently reported a method to fabricate freestanding LaAlO₃/SrTiO₃ (LAO/STO) membranes by spalling of strained heterostructures[1] and in Ref. [2] the first steps were reported towards controlling the spalling process towards uniform LAO/STO micromembranes with controlled sized in the range 2-20 μm. We discuss here a scheme for high-yield fabrication of membrane devices on silicon and show that the membranes exhibit metallic conductivity and a superconducting phase below ~200 mK. Using anisotropic magnetotransport we extract the superconducting phase coherence length $\xi \approx 36\text{--}80$ nm and establish an upper bound on the thickness of the superconducting electron gas $d \approx 17\text{--}33$ nm, thus confirming its two-dimensional character. Finally, we show that the critical current can be modulated using a silicon-based backgate. The ability to form superconducting nanostructures of LAO/STO membranes, with electronic properties similar to those of the bulk counterpart, opens opportunities for integrating oxide nanoelectronics with silicon-based architectures.



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Metal-to-Insulator Transition in Antiferromagnetic SrCrO₃ Thin Films

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Perovskite transition metal oxides are widely studied fascinating materials that have been the subject of numerous reports. This is especially true for the first-row transition metals, with the exception of the chromium family ReCrO₃ (Re being a divalent rare earth), which remains only rarely studied. This is mainly explained by its difficult synthesis, the stabilization of Cr⁺⁴ (characterized by two electrons in the nearly triply degenerate t_{2g} orbitals) typically requires high pressures [1, 2]. Among this family of materials, SrCrO₃ (SCO) has first been synthesized in 1967 [1] and its properties are still debated nowadays. It is suggested to be metallic and antiferromagnetic (AFM) in its ground state, a phase coexistence that is rare and poorly understood [3].

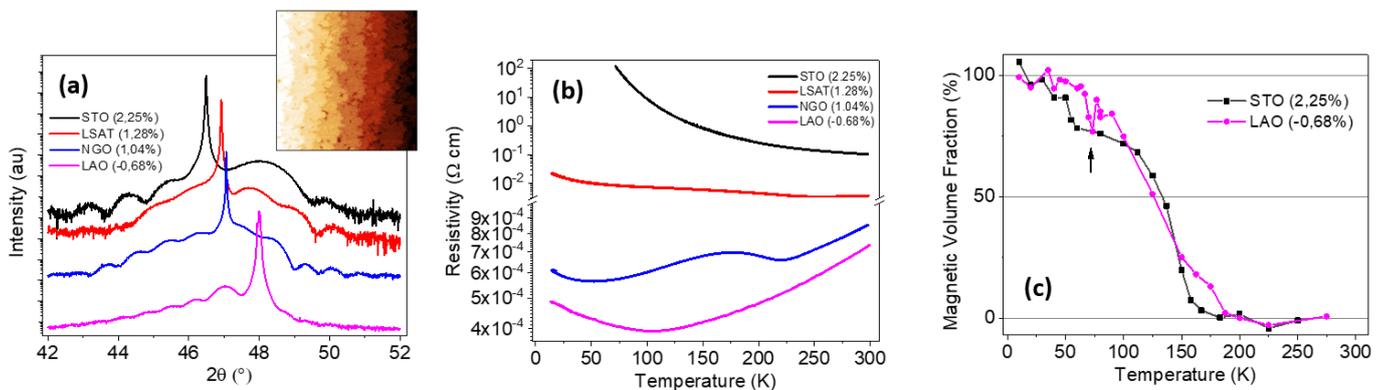


Figure 1: (a) X-ray diffraction measurements showing the high crystalline quality of SCO thin films grown on SrTiO₃ (STO), LSAT, NdGaO₃ (NGO) and LaAlO₃ (LAO). The strain values are indicated in the legend. The inset shows an atomic force microscope image of a SCO film grown on STO (the side-scale is 2 μm). (b) Resistivity measurements of SCO thin films. (c) Magnetic volume fraction measured by muon spin relaxation on a film grown on STO (black line) and LAO (magenta line).

Here we report the successful growth of high-quality epitaxial SCO thin films by off-axis RF magnetron sputtering. A very low but finite amount of oxygen was required to obtain the desired Cr⁺⁴ valence state in the films, as verified by x-ray absorption spectroscopy (XAS). The high crystalline quality thin films have been grown on a variety of substrates imposing a broad range of strains (from -0.68% to 2.25%, see Figure 1(a)). Transport measurements on SCO films have revealed a metal-to-insulator transition upon application of strain (Figure 1(b)). Under small compressive and tensile strain, SCO thin films show a metallic behaviour. As the tensile strain is further increased, a transition to an insulating behaviour is observed. This result, in agreement with previous work [4], is explained by DFT+U calculations [5]. A Jahn-Teller (JT) structural distortion is responsible for lifting the orbital degeneracy, hence opening a gap in the band structure. This JT distortion also appears to be compatible with a C-type AFM ordering. In order to directly probe the existence of a magnetic phase in SCO films, we performed muon spin relaxation experiments. These measurements have revealed a clear magnetic transition in SCO thin film at about 130 K. Moreover, this transition appears to be independent of strain (Figure 1(c)). These measurements represent the first direct observation of a metallic-AFM phase coexistence in SCO thin films.

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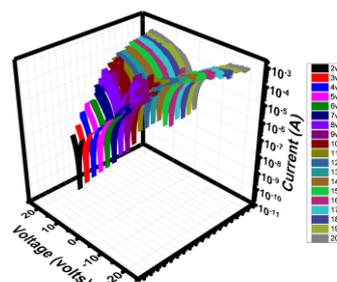
Low power resistive switching characteristics in Metal Oxide's for transparent neuromorphic devices

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Transparent neuromorphic electronics has attracted significant scientific attention for developing invisible circuits, ultra-low power electronic devices, sensors, and artificial synapses[1]–[4]. Data storage media have been actively studied; especially resistive switching memories are attracting attention as next generation memory devices. Therefore, reducing the power consumption will benefit the practical process for data storage and neuromorphic devices[5]–[7]. Another related concept is memristive behavior where the resistance value is continuously modified with the history of applied voltage and current as shown in figure. This mimics the situation in the brain, in which the electrical connections between two neurons become stronger every time the connection is addressed[8], [9]. The device has a very simple structure in which the resistance changing layer is sandwiched between upper and lower metal electrodes. Here, we have fabricated the amorphous oxide thin films on transparent conducting oxide coated 7059 glass substrates, using electron beam evaporation. To realize a low power RS/Memristive device, it is critical that the operating current should be as low as possible. Due to the presence of large defects in oxides (oxygen vacancies) and leakage current this is a daunting task to find the materials combinations. Some metal oxides such as Tantalum Oxide, Hafnium Oxide have got recent attention due to wide bandgap and high dielectric constants. We have found that the overall character of RS is different for different thickness layer. These results along with various structural and optical measurements will be presented and discussed.



Memristive behavior of transition metal oxide.

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Electric control of two-dimensional Van Hove singularity in oxide ultrathin films

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The divergent density of states (DOS) can induce abnormal phenomena such as large enhancement of superconductivity and unexpected phase transitions. Van Hove singularities (VHSs) are known to induce divergent DOS in two-dimensional (2D) systems. In spite of the recent high interest in the VHS, only a few controllable VHSs have been reported. Here, by utilizing an atomically ultrathin SrRuO₃ (SRO) film, we investigate the electronic band structures of the 2D VHS with angle-resolved photoemission spectroscopy (ARPES) and control the transport properties. By applying electric fields with alkali metal deposition and ionic-liquid gating methods, we precisely control the 2D VHS and the sign of the charge carrier. The tunable 2D VHS in an atomically flat oxide film can be a new strategy to realize infinite DOS near the Fermi level, thereby allowing us to engineer electric properties efficiently.

Quantum effects in the electronic properties of epitaxial SrTiO₃ films on Si(001)

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The two-dimensional (2D) electron systems at the interface between SrTiO₃ and oxides and on the surface of SrTiO₃ have been investigated intensely in the past years. SrTiO₃ and Si are two of the most well-studied systems in oxides and semiconductors, respectively. The tunability of their electronic properties constitutes them ideally for technological applications. SrTiO₃ can grow coherently on Si(001) by a 45° rotation around the surface normal using oxide Molecular Beam Epitaxy (MBE)[1].

We investigate the electronic properties of an oxide-semiconductor heterostructure, SrTiO_{3.8} thin films grown epitaxially on Si(001) using low-temperature magnetotransport measurements. We show that the temperature and magnetic field dependence of the conductivity is dominated by two-dimensional quantum effects, weak localization and electron-electron interactions. Quantum confinement, strong electron correlations, exchange interaction, and Zeeman spin-splitting are demonstrated, pointing to the realization of an oxide quantum well on Si.

For low carrier concentrations $<10^{12}$ cm⁻² epitaxial strain seems to strengthen electron-electron interactions while at higher electron concentrations $>10^{14}$ cm⁻² spin-orbit coupling dominates [2]. For both low and high electron concentrations we observe single-carrier transport. At intermediate electron densities $\sim 10^{13}$ cm⁻² (near the Lifshitz transition point) and below 40 K, two-carrier transport is evident by a non-linearity of the Hall resistance in the applied magnetic field [3]. This non-linear behavior is evaluated using a two-carrier model and can be accounted for by the presence of two distinct electron carriers of different concentrations and mobility. Temperature and magnetic field dependence measurements are used to evaluate the effects of quantum interference and electron-electron interactions on the conductivity.

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Room-Temperature Valence Transition in a Strain-Tuned Perovskite Oxide

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Cobalt oxides have long been understood to display intriguing phenomena known as spin-state crossovers, where the cobalt ion spin changes vs. temperature, pressure, *etc.* A very different situation was recently uncovered in praseodymium-containing cobalt oxides, where a *first-order, coupled* spin-state/structural/metal-insulator transition occurs, driven by a remarkable praseodymium *valence transition*. Such valence transitions, particularly when triggering spin-state and metal-insulator transitions, offer highly appealing functionality, but have thus far been confined to cryogenic temperatures in bulk materials (*e.g.*, 90 K in $\text{Pr}_{1-x}\text{Ca}_x\text{CoO}_3$). In this work we show that in thin films of the complex perovskite $(\text{Pr}_{1-y}\text{Y}_y)_{1-x}\text{Ca}_x\text{CoO}_{3-\delta}$, heteroepitaxial strain tuning (Fig. 1(a)) enables stabilization of valence-driven spin-state/structural/metal-insulator transitions to *at least 291 K, i.e., around room temperature* (Fig. 1(b)) [1]. The technological implications of this result are accompanied by fundamental prospects also, as complete strain control of the electronic ground state is demonstrated (Fig. 1(c)), from ferromagnetic metal under tension to nonmagnetic insulator under compression, thereby exposing a potential novel spin-state quantum critical point [1].

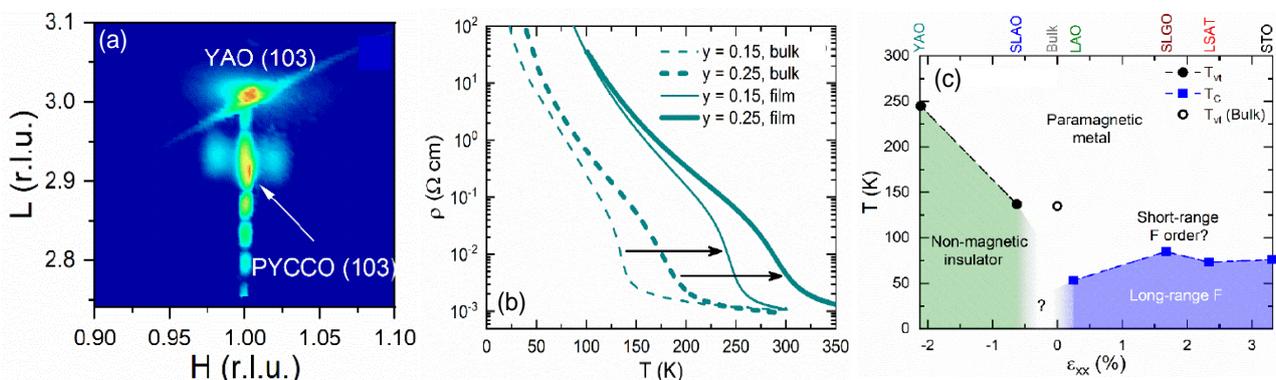


Figure 1: (a) Synchrotron X-ray reciprocal space map illustrating fully-strained growth of $(\text{Pr}_{0.85}\text{Y}_{0.15})_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ on YAlO_3 (2.1% compression). (b) Temperature-dependent resistivity of $(\text{Pr}_{1-y}\text{Y}_y)_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ with $y = 0.15, 0.25$ in bulk and film form. Note the enhancement of the transition temperature with strain (arrows), reaching 291 K for $y = 0.25$. (c) Strain “phase diagram” of $(\text{Pr}_{0.85}\text{Y}_{0.15})_{0.7}\text{Ca}_{0.3}\text{CoO}_{3-\delta}$ showing the valence transition (T_{vt}) and Curie (T_c) temperatures.

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Anomalous Hall effect in SrIrO₃ and SrRuO₃ thin films

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Heavy transition metal oxides represent an interesting playground for unusual magnetotransport behavior including anomalous Hall effect (AHE), topological Hall effect (THE), quantum anomalous Hall effect (qAHE) and spin Hall effect (SHE). Due to the interplay between crystal-field, short-range electron-electron correlations and strong spin-orbit coupling, they show various topological features in their electronic band structures, which are connected to their anomalous transport behavior. Among the exotic transport phenomena, AHE, which is induced by a large spin-orbit coupling, has been one of the most intensively studied so far. Recently, a strain-tuned pyrochlore iridate, Nd₂Ir₂O₇, epitaxially grown on YSZ substrates, has been reported to exhibit a large AHE by manipulating the Berry curvature in this spin-orbit coupled antiferromagnet [1]. It also has been recently demonstrated that the non-polar/polar interface of a SrRuO₃ film with a LaAlO₃ (LAO) capping layer shows a sign change of the AHE depending on the LAO thickness [2].

In this poster, we report on the observation of the AHE in two oxide heterostructures, SrIrO₃/SrTiO₃(111) and LaAlO₃/SrRuO₃/SrTiO₃(100). We investigate their high-quality crystal structures using transmission electron microscopy and correlated photoemission data on both core-levels and valence band with the findings of the transport measurements. We also discuss the relationship between spin configurations and Berry curvature.

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Thickness influence of SrTiO₃ buffer on CNO nanosheet templated PZT for High electron mobility transistor devices

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High band gap semiconductors possessing high breakdown fields are in strong demand for high power and high frequency applications. Gallium nitride (GaN) with a wide band gap therefore a promising candidates for such devices [1]. The integration of ferroelectric materials such as Pb(Zr_{0.52}Ti_{0.48})O₃ (PZT) offers a strong potential for improving properties like larger breakdown voltage and better on/off ratios [2]. A large lattice mismatch between GaN and PZT however poses a challenge for epitaxial growth. The integration of Ca₂Nb₃O₁₀ (CNO) nanosheets able to function as a template for epitaxial growth of PZT qualifies as a possible solution[3]. Previous research on CNO nanosheets templated PZT showed that a thin perovskite oxide buffer layer could enhance the properties significantly [4]. SrTiO₃ (STO) showed most prominent improvements and is thus further studied in this work. The influence of the STO buffer thickness, varying from 1-25nm, on the PZT properties and the interfacial effects playing an important role in the system are investigated.

CNO nanosheets are deposited using the Langmuir-Blodgett technique. Thin films of STO and PZT are grown by pulsed laser deposition (PLD). AFM, XRD and SEM are used for characterizing the topography and the crystallographic properties of the PZT films. It can be concluded that a thin STO buffer already significantly enhances the crystallographic as well as the electrical properties of PZT.

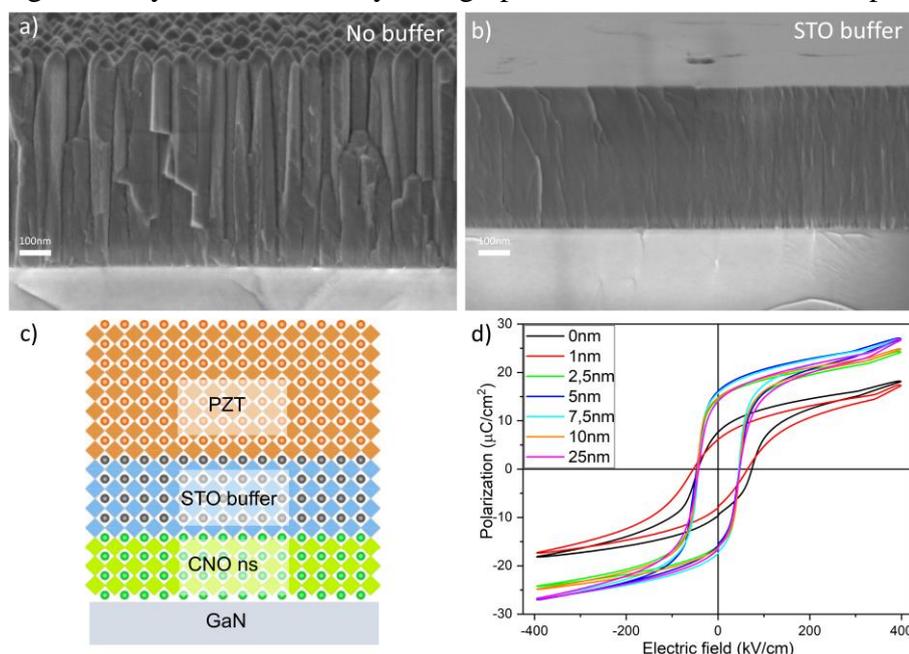


Figure 1: Cross-sectional SEM of PZT on CNO ns using no buffer and an STO buffer, a) & b) respectively. Schematic representation of grown films on GaN, c). Polarization hysteresis of PZT using different buffer thicknesses, d).

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Higher harmonics in planar Hall effect induced by cluster magnetic multipoles

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Antiferromagnetic (AFM) materials are attracting tremendous attention due to their spintronic applications [1] and associated novel topological phenomena [2]. However, detecting and identifying the spin configurations in AFM materials is quite challenging due to the absence of net magnetization. Herein, we report the practicality of utilizing the planar Hall effect (PHE) to detect and distinguish “cluster magnetic multipoles [3]” in AFM Nd₂Ir₂O₇ (NIO-227) fully strained films. By imposing compressive strain on the spin structure of NIO-227, we artificially induced cluster magnetic multipoles, namely dipoles and A₂- and T₁-octupoles. Importantly, under magnetic field rotation, each magnetic multipole exhibits distinctive harmonics of PHE oscillation. Moreover, the planar Hall conductivity has nonlinear magnetic field dependence, which can be attributed to the magnetic response of the cluster magnetic octupoles. Our work provides a strategy for identifying cluster magnetic multipoles in AFM systems, and it would promote octupole-based AFM spintronics.

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Unconventional anomalous Hall effect in Ru(III) perovskite oxide thin films

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Perovskite ruthenates ARuO_3 are one of the most typical conducting complex oxides. Previous efforts on ARuO_3 , however, have been mostly focused on Ru^{4+} compounds ($\text{ARu}^{4+}\text{O}_3$), while there have been only a few reports on ARuO_3 with Ru^{3+} ($\text{ARu}^{3+}\text{O}_3$) [2,3]. In contrast to the well-studied $\text{ARu}^{4+}\text{O}_3$, $\text{ARu}^{3+}\text{O}_3$ can accommodate Ln^{3+} (Ln : lanthanides) on A -site, which can render this system an intriguing platform for emerging magnetotransport properties because of the interaction between Ru - $4d$ itinerant electrons and Ln - $4f$ localized spins. In this study, by fabricating single crystalline thin films of LaRuO_3 and NdRuO_3 , we demonstrate the difference of their magnetotransport properties and successfully capture the unconventional anomalous Hall effect (AHE) in NdRuO_3 .

LaRuO_3 and NdRuO_3 thin films are fabricated on SrTiO_3 (001) substrates by pulsed laser deposition and subsequent annealing. Through magnetotransport measurements up to 14 T and down to 50 mK, LaRuO_3 is found to possess a paramagnetic metallic ground state. In a stark contrast, for NdRuO_3 , temperature dependence of longitudinal resistivity is metallic at higher temperatures and shows an upturn at ~ 23 K. Below 10 K, negative magnetoresistance and significantly non-linear Hall effect are observed. To elucidate the origin, further magnetotransport measurements are performed up to 54 T. It is revealed that Hall coefficient shows almost constant negative value at higher field region below ~ 150 K (Fig. 1(a)), with which anomalous Hall resistivity (ρ_{AHE}) is deduced. AHE starts to emerge at ~ 120 K and additional AHE term develops below ~ 30 K (e.g., see $B = 1$ T in Fig. 1(b)). These features are possibly indicative of the contribution from slowly-saturated Ru^{3+} and quickly-saturated Nd^{3+} moments, respectively. Moreover, below ~ 2 K, an unconventional hump structure is observed in magnetic field dependence of ρ_{AHE} at ~ 1 T (Fig. 1 (c)). In consideration of the $g_y a_x$ type anti-ferromagnetic (AFM) order of Nd^{3+} moments commonly observed in other NdBO_3 compounds [4], this unconventional AHE plausibly originates from the topological origin of non-coplanar spin textures configured during the magnetization process from the AFM to an induced ferromagnetic states.

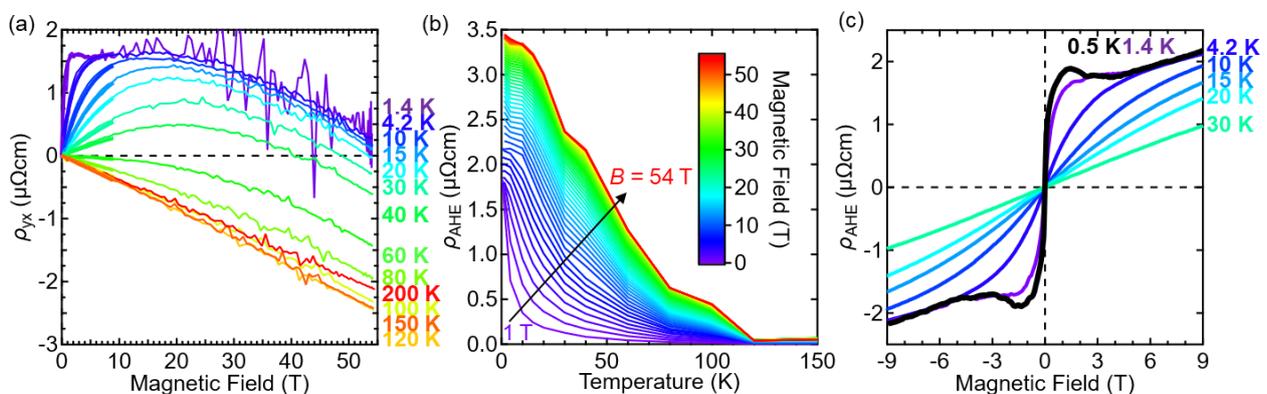


Figure 1 (a) Magnetic field dependence of Hall resistivity (ρ_{yx}) at various temperature up to 54 T and down to 1.4 K. (b) Temperature dependence of anomalous Hall resistivity (ρ_{AHE}) at various magnetic field. (c) Magnetic field dependence of ρ_{AHE} at low field region down to 0.5 K.

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Correlated Electron Metals with High Entropy

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Novel conducting complex oxides with high electron correlation are of keen interest for UV-transparent applications, for quantum spin devices, as high energy plasmonic platforms, and as hosts for fundamental studies of correlated electron systems. The family of Sr based oxide perovskites, including Sr_xVO_3 , Sr_xNbO_3 and Sr_xMoO_3 , is an interesting family to explore due to their heavy cation solubility and tunable conductivity and transparency. Stabilizing this family in the perovskite phase is required to harness the conflicting combination of optical and carrier transport properties. Transparency originates from the energetic isolation of the conduction band from the transition metal d-orbitals and the valence band from oxygen 2p-orbitals in the perovskite phase. Utilizing a high configurational entropy formulation, the high symmetry perovskite phase can be stabilized with five cations on the perovskite B-site. The high entropy approach expands the materials design space of transparent conductors beyond conventional enthalpy-stabilized phases. This enables a new materials design scheme that leverages local disorder to enhance optical transparency by suppressing interband transitions through state occupancy engineering and enhance conductivity by populating t_{2g} d-orbitals with conduction carriers. In this work, $\text{Sr}_x(\text{Nb},\text{Mo},\text{Ti},\text{W},\text{Cr})\text{O}_3$ and its derivatives were prepared and investigated by pulsed laser deposition and electrical transport analysis. Particular attention is paid to the relationship linking target formulation, synthesis conditions, and substrate crystal structure to the overall ability to stabilize perovskite vs. other phases. X-ray diffraction and atomic force microscopy results will be presented to demonstrate condition/formulation combinations that produce perovskite films and those which promote step-flow growth. Transport properties are characterized by temperature dependent resistivity analysis, ellipsometry and UV-Vis analysis. Cation distribution and oxidation states are studied by energy dispersive X-ray spectroscopy and X-ray photoelectron spectroscopy. First principles calculations are performed to study the effect of the coexistence of cations on the band structure and orbital filling. While the derivative systems of $\text{Sr}_x(\text{Nb},\text{Mo},\text{Ti},\text{W},\text{Cr})\text{O}_3$, with four B-cations instead of five, exhibit chemical and/or phase segregation, our results show that $\text{Sr}_x(\text{Nb},\text{Mo},\text{Ti},\text{W},\text{Cr})\text{O}_3$ thin films can be stabilized as single-phase perovskite. Contributions from individual B-cations and the interactions amongst them result in the unique overall properties of $\text{Sr}_x(\text{Nb},\text{Mo},\text{Ti},\text{W},\text{Cr})\text{O}_3$. For instance, Cr dramatically influences structure, crystallinity, oxidation, and transport. Additionally, A-site vacancies influence structure stability and electronic properties.

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Mg²⁺ diffusion across epitaxial Fe₃O₄/MgO(001) interfaces

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Epitaxial Fe₃O₄ thin films grown on single crystal MgO(001) present well-defined model systems to study oxide electronics and spintronics. However, the interface and film properties can be easily affected by the facile Mg²⁺ ion diffusion processes. In this work, we show at an atomic scale the Mg²⁺ diffusion pathways, kinetics, and reaction products at the Fe₃O₄/MgO heterostructures under different oxygen partial pressures and thermal annealing conditions. Combining microscopic, optical, and spectroscopic techniques, we demonstrate that an oxygen rich environment promotes Mg²⁺ incorporation into the Fe²⁺ sites, leading to the formation of Mg_{1-x}Fe_{2+x}O₄ spinel structures, where the corresponding portion of the Fe²⁺ ions are oxidized to Fe³⁺. On the other hand, annealing in vacuum results in the formation of a thin interfacial rock-salt layer (Mg_yFe_{1-y}O), which serves as a blocking layer leading to significantly reduced Mg²⁺ diffusion to the bulk Fe₃O₄. The roles of Ca impurities in MgO and defects in Fe₃O₄ will also be discussed. Our results reveal the critical role of available anions in governing cation diffusion in the spinel structures, and the need to prevent formation of unwanted reaction intermediates for the promotion of facile cation diffusion.