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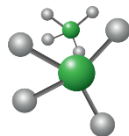


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Thin film epitaxy of low valence metallic oxides: from negatively valent pnictides to divalent rare earth monoxides

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In this talk, we show thin film epitaxy of a layered oxide R_2O_2Pn and rocksalt RO (Pn : Sb and Bi; R : rare earths).

A layered oxide R_2O_2Bi with anomalously low valent Bi^{2-} square net shows interesting electronic properties such as metal-insulator transition and superconductivity [1,2], although it is difficult to grow the single crystal. Direct thin film growth of R_2O_2Bi is also difficult, resulting in the formation of much more stable $R_2O_3\cdot Bi$. Thus, multilayer solid phase epitaxy was developed to obtain highly crystalline Y_2O_2Bi epitaxial thin film by using multilayer precursor $[Bi/Y/Bi/Y/Y_2O_3]_n$ grown by sputtering [3,4]. By using this approach, highly conducting La_2O_2Sb and semimetallic La_2O_2Bi epitaxial thin films were obtained in contrast with the bulk specimens [5,6].

Sesquioxide R_2O_3 with very stable trivalent rare earth ion is the most stable rare earth binary oxide, which is highly insulating [7]. On the other hand, several light rare earth monoxides RO with divalent rare earth ions were synthesized only many years ago [8]. Thin film epitaxy with pulsed laser deposition was found to be useful to obtain single crystalline rocksalt type YO [9]. Subsequently, epitaxial thin films of LaO , CeO , PrO , NdO , SmO , GdO , TbO , HoO , YbO , and LuO were successfully obtained [10]. These RO are highly conducting, and some are ferromagnetic or superconducting, in stark contrast with R_2O_3 , reflecting $4f^m5d^1$ electronic configuration of the divalent R ions. It is noted that heavy RO possesses T_C significantly higher than that of EuO (69 K). Among them, GdO has the highest T_C of 303 K by improving the crystallinity [11].

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Superconductivity at the KTaO_3 interfaces

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2D superconductivity is a fertile landscape for exotic quantum phenomena. Rashba and Gor'kov suggested that the interfaces with broken inversion symmetry could be an ideal platform for unconventional 2D superconductivity, including finite momentum and triplet pairing, with enhanced critical field beyond Pauli limit [1]. The nature of superconductivity and its interplay with strong spin-orbit coupling at the $\text{KTaO}_3(111)$ interfaces remain a subject of debate. This talk will focus on our recent experiments of epitaxial growth [2] and superconductivity [3] of KTaO_3 heterostructures. Superconductivity at KTaO_3 interfaces is robust against the in-plane magnetic field, with the critical field of superconductivity reaching ~ 25 T in optimally doped heterostructures [4]. The superconducting order parameter is highly sensitive to the carrier density and orientation of the magnetic field. The strong violation of Pauli limit for the upper critical field of the KTaO_3 interfaces is due to spin-orbit coupling which motivates the formation of anomalous quasiparticles with vanishing magnetic moment. These results offer design opportunities for superconductors with extreme resilience against the applied magnetic fields.

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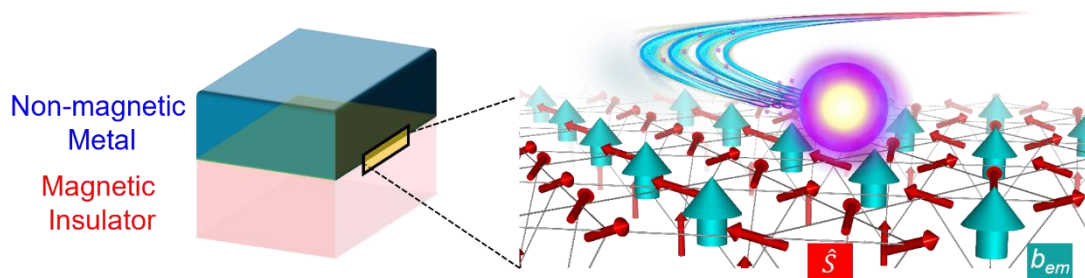
Emergent magnetotransport phenomena at oxide interfaces

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Oxide interfaces have been the objects of interest in condensed matter physics as the mesmerizing materials platform for emergent phenomena [1]. Entanglement of multiple degrees of freedom at the interfaces leads to numerous emergent physical properties not found in the bulk constituents. Another rapidly growing field is topological electromagnetism due to the non-trivial topology of electrons either in real (spin) or reciprocal (band) spaces, giving rise to diverse emergent magnetotransport phenomena. The topological Hall effect (THE) is one of the most well-known manifestations and is rooted in the “emergent field” originating from a non-coplanar spin texture with a finite scalar spin chirality. Integration of these research fields resulted in the discovery of THE at oxide interfaces [2] as well as the arguments about its origin.

Motivated by these developments, we have been pursuing the possibility of realizing THE at oxide interfaces composed of a non-magnetic metal and a magnetic insulator to harness this fascinating transport phenomenon. In this talk, we will present our recent studies on the interfacial emergent magnetotransport phenomena in oxide heterostructures driven by the “proximity effect of the emergent field”. Starting from the initial proof of concept in $\text{Bi}_2\text{Rh}_2\text{O}_7/\text{Dy}_2\text{Ti}_2\text{O}_7$ [3], we have observed THE with an effective field as large as ~ 30 T in $\text{CaRuO}_3/\text{DyFeO}_3$ [4]. Our findings offer significant advantages in both fundamental and applied physics by expanding the range of materials beyond conventional magnetic metals with non-trivial spin textures. We believe our approach will chart a new pathway to the practical application of the emergent field, building on the decades-long history of oxide electronics.



Concept of the interfacial emergent magnetotransport phenomena.

A conducting electron in a non-magnetic metal is driven by the emergent field (b_{em}) generated in a magnetic insulator with non-trivial spin texture (\hat{S}) hosting scalar spin chirality.

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Atomic-scale insights to lattice and electronic structure in superconducting nickelates

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The superconducting nickelates are a premier example of how breakthrough advances in materials synthesis and characterization techniques over the past decades have enabled the realization and discovery of new materials families [1]. Within the phase space of notoriously challenging synthesis [2,3], many materials realities including epitaxial strain, extended defects, impurities, and secondary phases have been as yet largely unexplored in the context of understanding their impact on the measured properties of these materials. With the ability to directly probe both lattice and electronic structure down to the atomic scale, scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) offer unique insights to build a holistic understanding of both the intrinsic physical properties in these materials as well as more extrinsic features and their consequences, many of which arise from their multistep synthesis. Here, we discuss how quantitative lattice-scale measurements can disentangle many of these effects. Through selective local spectroscopy we reveal, for example, the Mott-Hubbard electronic landscape and multi-band hole interactions across the hole-doped phase diagram in infinite-layer nickelates [4]. We also investigate the electronic impact of a nontrivial interface reconstruction through close iterative feedback between quantitative experimental measurements and explicit large-cell density functional theory calculations [5]. We further systematically explore the role played by — and limits to — epitaxial strain in these systems [6, 7]. Ultimately, these insights contribute to dramatic synthetic improvements to realize high-quality samples suitable for a range of fundamental measurements.

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Engineering ultrafast dynamics and non-equilibrium phases in atomically layered oxides

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One of the key approaches to manipulating functionalities in oxide materials has been to make use of atomically layered heterostructures, where epitaxial strain, dimensional confinement, octahedral connectivity, and charge transfer can be employed. This approach has been widely applied to manipulate the static properties of oxide materials in thermal equilibrium, but little work has explored the use of such heterostructuring to control the dynamic, non-equilibrium behavior of such systems. In this talk, I will describe recent experiments in which we demonstrate the ability to tune the ultrafast dynamics of optically driven oxides by engineering their structure and dimensionality at the atomic scale. For example, I will show results in which intertwined orders can be decoupled enabling the formation of “hidden” charge, spin, and structural phases and illustrate that epitaxial and light- induced strains can be combined to stabilize highly non-equilibrium ground states, including a room- temperature multiferroic phase. These experiments expose a new budding “non-equilibrium materials design” paradigm for the creation of desirable dynamical functionalities in oxide electronics.

Revisit the magnetotransport of perovskite manganite heterostructures

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In transition-metal-oxide heterostructures, the anomalous Hall effect (AHE) serves as a powerful tool for detecting magnetic states and revealing intriguing interfacial magnetic orderings. However, achieving a larger AHE at room temperature in oxide heterostructures remains challenging due to the dilemma of mutually strong spin-orbit coupling and magnetic exchange interactions. In this study, we use perovskite manganite heterostructures as a model system and employ several experimental approaches to enhance the magnetotransport signal. First, we explored the Ru doping-enhanced AHE in $\text{La}_{2/3}\text{Sr}_{1/3}\text{Mn}_{1-x}\text{Ru}_x\text{O}_3$ epitaxial films. By increasing the B-site Ru doping level up to 20%, we observed an enhancement in anomalous Hall resistivity at room temperature from $\text{n}\Omega\cdot\text{cm}$ to $\mu\Omega\cdot\text{cm}$ scale. Ru doping leads to strong competition between ferromagnetic double-exchange interaction and antiferromagnetic super-exchange interaction. The resultant spin frustration and spin-glass state facilitate a strong skew-scattering process, thus significantly enhancing the extrinsic AHE [1,2]. Second, we discovered in-plane AHE (IP-AHE) in the ruthenate/manganite/ruthenate trilayer heterostructures. Both the ruthenate capping/buffer layers effectively modulate the MnO_6 octahedral rotation/distortion pattern of the manganite layer, thereby resulting in an in-plane uniaxial magnetic anisotropy. The simultaneous breaking of inversion and time-reversal symmetries induce a pronounced IP-AHE, reaching up to $100 \text{ n}\Omega\cdot\text{cm}$. Moreover, the IP-AHE can be effectively modulated by various stimuli, including epitaxial strain, interfacial structure, and magnetic field [3]. At last, we demonstrate the tunable magnetic skyrmions and the corresponding topological Hall effect of manganite thin films.

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New Directions in Material Design: Harnessing Freestanding Oxide Membranes

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Complex oxides are crucial in advanced electronics, energy, and information technologies due to their diverse functionalities, including ferroelectricity, piezoelectricity, and pyroelectricity. These properties arise from the complex interactions among lattice, orbital, charge, and spin dynamics within the materials. In this presentation, I will discuss recent advancements in manipulating the properties of oxide heterostructures by deliberately altering their symmetry. I will particularly focus on the emerging area of freestanding oxide membranes. These membranes, mainly made from transition metal oxides, are thin, quasi-2D layers that have become essential for experimentalists aiming to develop materials with novel properties. They are engineered into multilayer structures where the twist angles between layers are precisely controlled. Additionally, I will introduce a novel approach for creating high-quality complex heterostructures integrated with Si using SrTiO₃ membranes. This method offers a new avenue for developing electronic devices that combine multifunctional physical properties with silicon technology.

From Oxide Epitaxy to Membranes: Opportunities and Challenges

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With a rapidly growing family of vdW materials, the role of dielectric and metals have become more important than ever. In this talk, I will present challenges associated with the synthesis of atomically-precise three-dimensional (3D) perovskite nanomembranes followed by our group's effort to address them. Using hybrid molecular beam epitaxy that employs a metal-organic precursor, titanium isopropoxide (TTIP), to supply both Ti and oxygen (without the need for additional oxygen), epitaxial SrTiO₃ (STO) films were grown directly on a graphene layer transferred on to bulk STO substrate. Films were then successfully exfoliated and transferred onto other substrates. Using Raman spectroscopy and high-resolution X-ray diffraction, we show that the transferred STO membrane is single-crystalline and can be integrated with other vdW materials. I will also present sacrificial layer route to create oxide membranes resulting in room temperature dielectric constant of ~ 300. Finally, I will present several opportunities for materials physics and devices engineering using 3D nanomembranes.

Epitaxial ferroelectric HfO₂: effects of stress and strain

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Ferroelectric HfO₂ films are mostly studied in their polycrystalline form. The mixture of polar and nonpolar phases with different orientations challenge the understanding of ferroelectric properties and impedes fine tuning of the ferroelectric orthorhombic phase stabilization. Several factors such as dopants, oxygen vacancies, or stress, among others, have been investigated and shown to have a crucial role on optimizing the ferroelectric response. Stress generated during deposition or annealing of thin films is a main factor, which determines the formed crystal phases and influences the lattice strain of the orthorhombic phase. It is difficult to discriminate between stress and strain effects in polycrystalline ferroelectric HfO₂ films, and it has been elusive to determine experimentally the direct impact of orthorhombic lattice strain on ferroelectric polarization. Due to that, our knowledge on ferroelectric fluorites is much less than the one on conventional ferroelectric perovskites.

Epitaxial layers are well suited to increase our understanding of ferroelectricity in fluorites [1]. We have investigated a wide range of epitaxial thin films with different doping and deposition parameters to reveal the governing mechanisms for achieving robust ferroelectric polarization. Correlations are established between polarization and orthorhombic phase content and orthorhombic lattice tension [2]. The content of the orthorhombic phase determines the polarization, while there are not evidences of a strong direct effect of the strain, which is confirmed by density functional theory calculations.

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Epitaxial oxides for neuromorphic devices

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Devices that emulate the behavior of neurons and synapses for extremely efficient information processing are becoming increasingly interesting. However, the number of materials that have been considered as the key elements in such devices is still restricted to a few materials that are compatible with CMOS integration. The need to limit the processing to low temperatures, often produces polycrystalline or amorphous phases that hamper the control of the material, reduce the device endurance and device-to-device reproducibility and obscure the understanding of the physical mechanisms at play. Thus, we propose that in the context of enabling brain-inspired, neuromorphic and in-memory computing, venturing into epitaxial, single-crystalline materials should cease to be seen as a waste of resources but rather as a way to find a direct access to the key materials issues and their solution. Emergent efforts towards epitaxial growth of complex oxides by ALD [1] allow us to dream of a future of epitaxial oxide microelectronics for which we want to be prepared. In this talk, we will show two examples of epitaxial materials showing interesting neuromorphic features that, to our knowledge, have not been reported in other materials: the particular self-oscillating behavior of epitaxial TbMnO_3 [2] as a compact neuristor [3] and the switchable behaviour between synapses and neurons demonstrated by ferroelectric/nickelate heterostructures [4].

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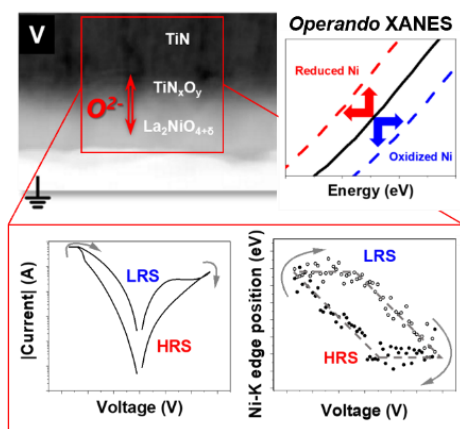
Operando spectroscopic investigation of the valence change mechanism in $\text{La}_2\text{NiO}_{4+\delta}$ - based memristive devices

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Resistive switching (RS) devices have attracted increasing attention for their application as non-volatile memories and neuromorphic computing systems. In my talk, I will focus on the design of novel memristive heterostructures using a perovskite-related oxide with high oxygen mobility and p-type semiconducting electronic transport, namely $\text{La}_2\text{NiO}_{4+\delta}$ (L2NO4). While our first studies were dedicated to epitaxial planar devices on single-crystal substrates [1-2], we then moved towards industrial-relevant substrates and thus fabricated vertical devices with polycrystalline L2NO4 films on Si-based CMOS-compatible wafers [3]. The electrical response of these devices could be tuned both by changing the electrode materials and the amount of point defects (oxygen interstitials) of the sandwiched film. An analog bipolar counter-eightwise RS behavior with multiple resistance states was proven using continuous sweeps and voltage pulses. The key role played by the oxygen interstitials on the initial resistance state, the initialization step, and the memristive characteristics of the devices will be presented and discussed.

Selected devices were further analyzed by X-ray absorption spectroscopy (XAS) using synchrotron radiation. [4] Changes in the Ni K-edge were measured for different annealings, in different regions of the device and under *in situ* and *operando* conditions, allowing for a better understanding of the RS mechanisms taking place in the device. Furthermore, gradual changes in conductance after the application of repetitive DC sweeps, which can be regarded as the evolution of the synaptic weight between neurons, were obtained for these novel L2NO4-based devices. These results were complemented with *in situ* transmission electron microscopy (TEM) measurements combined with Electron Energy Loss Spectroscopy (EELS) carried out for “lamella-devices” using an adapted TEM sample holder, which allowed for the observation of chemical changes for different resistance states. These results go a step further towards the comprehension of the intricate switching mechanisms taking place, and open the door to the use of L2NO4-based memristors as artificial hardware synapses in spiking neural networks [4].



Original operando XANES methodology based on monitoring the absorbance intensity changes at a fixed energy position

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Strange magnetism of orthorhombic iridates

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A complex interplay of correlations, spin-orbit coupling and lattice structure has acquired a prominent place in solid-state research in recent years, and the materials that are characterized by novel electronic, magnetic and topological properties due to a fine balance of these ingredients have loosely been coined “quantum materials”. *5d*-transition metal oxides, where correlations and spin-orbit coupling meet at the same energy scale, create a great playground in search for exotic physics, such as high- T_C and unconventional superconductivity, spin liquids, formation of polarons and novel magnetic ordering phenomena.

Iridium compounds, in particular, have received a lot of attention in the recent years. In the Ruddlesden-Popper series, $\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1}$, for example, one could investigate the spin-orbital interactions leading to the formation of the relativistic $j_{\text{eff}}=1/2$ state and a Mott gap, follow the changes of the correlation strength, the size of the gap, and the magnetic ordering for the different structures defined by the number of the perovskite units, n , within the materials’ unit cells [1]. The corresponding physical properties have been observed to change between an antiferromagnetic relativistic Mott insulator for $n=1$ to a nonmagnetic Weyl semimetal for $n = \infty$, offering the opportunity to understand the impact of the relativistic phenomena and correlations on the electronic transport and magnetic behavior, all within the same series.

The role of the Sr-cation in these iridates was investigated by its gradual substitution by Ca, reaching the metastable perovskite phase of CaIrO_3 as the end-point, in thin-film form [2]. The CaIrO_3 films were characterized as semimetallic by *ab-initio* calculations, although the optical conductivity spectra were inconclusive in this respect, leaving the possibility of a small-gap insulating state open, while the later synthesized bulk material was, as in the case of SrIrO_3 , found to be semimetallic and nonmagnetic [3].

We present our study of CaIrO_3 in the form of thin films grown on SrTiO_3 , employing a combination of *ab-initio* tools in combination with the resonant inelastic X-ray scattering. The results indicate that the films likely lie in the antiferromagnetic semimetallic regime. Ordering of higher-order magnetic multipoles is shown to go hand-in-hand with this peculiar state. We will discuss the possible consequences of this finding in connection to the structural and transport properties of the material.

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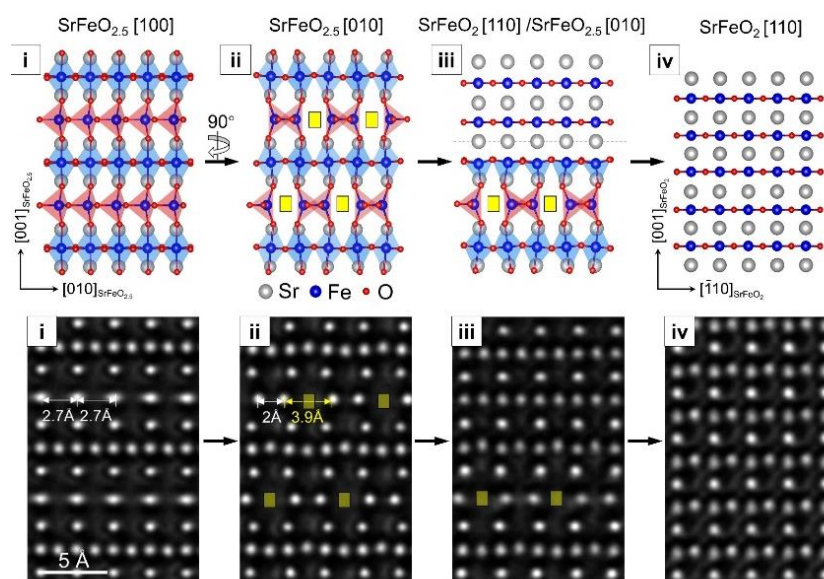
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Topotactic phase transformation in ferrites: octahedral, tetrahedral, and square planar FeO_x configurations

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Oxygen plays an essential role in determining the materials properties of transition metal oxides. By changing the oxygen concentration only by $\sim 30\%$, the electronic ground state of complex oxides may vary from insulator to metal to superconductor. Despite the general lattice structural similarity offered by the topotactic phase transformation, the change in the oxygen content necessarily accompanies the surrounding electronic environment of the transition metal – oxygen polyhedra, and the dimensionality and anisotropy of the polyhedral network. This largely modifies the electronic and phononic energy band dispersion along with the change in the apparent valence state of the transition metal ions. Hence, after the discovery, the topotactic phase transformation has been frequently employed to explore novel hidden physical properties with metastable structures. A real-time atomic-scale observation of the topotactic phase transformation, if plausible, would provide microscopic understanding of the dynamic evolution of the oxygen removal of conventional perovskite oxides, from octahedral to tetrahedral to square planar polyhedral configurations. Here, we present a prototypical example of such evolution in SrFeO_x epitaxial thin films. In particular, the anisotropic brownmillerite structure promote unexpected sequential structural evolution associated with the facile movement of the oxygen within the structure. Our study would help construct a design principle in obtaining novel oxide phases with diverse transition metal – oxygen polyhedral configurations.



Real-time atomic-scale observation of the topotactic phase transformation toward infinite-layer SrFeO_2 .

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<p>Ion irradiation induced surface electronic structure variation in the epitaxial lanthanum nickelate based heterostructures Sharma, Vishal (Micron Semiconductor Technologies, India)</p>
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Electron-beam writing of atomic-scale reconstructions at oxide interfaces

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Transition metal oxides exhibit a variety of functionalities including ferroelectricity, ferromagnetism, and high-temperature superconductivity. Epitaxial growth enables the synthesis of high-quality films, yet it confines the choice of growth substrates to those meeting symmetry and lattice parameter constraints, thereby limiting technological applications of epitaxial oxides. To overcome these constraints, oxide layers can be released from their growth substrate using epitaxial sacrificial layers positioned between the substrate and the film [1]. This approach allows the fabrication of oxide membranes, enabling the creation of novel heterostructures by non-epitaxial stacking of membranes and opening new possibilities for materials design.

Here, we report the controlled formation of interfacial ionic bonds between a 30 nm-thick SrTiO₃ (STO) membrane and a niobium-doped SrTiO₃(001)-oriented (Nb: SrTiO₃) carrier substrate [2]. The STO membranes were fabricated by growing a Sr₃Al₂O₆ (SAO) sacrificial layer and a 30 nm-thick STO layer on a STO(001) substrate using pulsed laser deposition. The SAO layer was dissolved in deionized water, and the resulting STO membrane was transferred onto the Nb: STO(001) carrier substrate. The crystal structure of the membrane before and after lift-off of the sacrificial layer was probed by X-ray diffraction, which confirmed that the crystallinity was preserved after release. Scanning transmission electron microscopy in electron energy-loss spectroscopy mode (STEM-EELS) was employed to investigate the electronic/bonding states of titanium (Ti) and oxygen (O) across the interface from the carrier substrate to the membrane, as a function of the annealing temperature. Our findings demonstrate that, after annealing, the STEM primary electron beam induces the rearrangement of interface atoms, which leads to a perfect interface reconstruction and the formation of ionic bonds between the membrane and the carrier substrate. STEM-EELS analysis showed that, after annealing and e-beam rastering, a change of Ti valency from Ti²⁺ to Ti⁴⁺ occurred and oxygen octahedral coordination was restored, indicating a crystal structure compatible with STO.

This study presents a method to selectively create ionic bonds between perovskite oxides, enabling the local manipulation of physical properties at their interface. Precise control of interface reconstruction between perovskite oxides therefore represents a powerful tool for discovering new interfacial phenomena, for instance by inducing tailored strain gradients. Our method suggests an alternative pathway for creating synthetic oxide membrane-based heterostructures, with the ability to selectively induce ionic bonding between them.

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Epitaxial VCrO₃ superlattice thin films on sapphire

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Corundum oxide thin films with formula M₂O₃ (space group R-3c) have been extensively investigated both for fundamental studies and their potential applications in oxide electronics. Examples include V₂O₃, Cr₂O₃, Fe₂O₃ and Ga₂O₃ which have been proposed for neuromorphic computing, magnetoelectric memories, supercapacitors and power electronics, respectively [1-4]. Their broad spectrum of physical properties can be further expanded by combining two or more oxides in a superlattice structure, taking advantage of the layered fashion of the corundum unit cell along the c-plane {001} or the r-plane {1-12}.

In this work we have grown VCrO₃ superlattices on c-plane sapphire substrates by molecular beam epitaxy (MBE). In-situ reflection high energy electron diffraction (RHEED) shows the periodic oscillations characteristic of a layer-by-layer growth. Structural characterization is then performed by X-ray diffraction and scanning transmission electron microscopy (STEM), including elemental mapping by energy-dispersive X-ray spectroscopy (EDX) and electron energy loss spectroscopy (EELS). Both X-ray reflectivity (XRR) and STEM confirm the successful growth of the superlattice thin films, featuring periodic V₂O₃ and Cr₂O₃ layers in the sub-unit cell regime, i.e. each thinner than 6 layers of cations. If compared to corundum, an additional A_g Raman active mode at 680 cm⁻¹ is observed, which has been linked to the symmetry breaking at the interfaces of a superstructure in ilmenite crystals (space group R-3) by Baraton et al. [5]. Ilmenite VCrO₃ is predicted to be thermodynamically stable by density functional theory (PBEsol+U) and it is expected to be a ferromagnetic insulator, in agreement with the calculation of Zhao et al. [6]. The insulating behavior is confirmed by 4-point probe resistivity measurements as a function of temperature for all the superlattice thin films. The magnetic properties of ilmenite VCrO₃ thin films are measured by superconducting quantum interference device (SQUID) magnetometry, revealing a weak ferromagnetism response at low fields of uncertain nature and no ferromagnetic transition up to 340K.

The successful MBE growth of VCrO₃ superlattice thin films in the sub-unit cell regime encourages further exploration of corundum-derived ABO₃ structures with the aim of exploring novel functional oxides as predicted by calculations [6, 7].

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Magnetic and polar properties of (010) DyFeO₃ thin films

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Growth induced strain in thin films is a means to modify materials properties since the sign of strain affects the bond length and therefore directly orbital interactions. Orthoferrites ($R\text{FeO}_3$) is one class of materials where interaction energies are small and strain modified properties are very prominent. DyFeO₃ as a representative of this family is a G-type antiferromagnet below 645 K in bulk with a spin reorientation transition of the Fe-spins between 40 and 50K and Dy ordering below 5 K. Due to a tilting of the oxygen octahedra along the c -axis, a weak ferromagnetic component is introduced which vanishes below the spin reorientation due to a change in magnetic symmetry. Applying a magnetic field of 2.3 T below the Dy ordering temperature, DyFeO₃ becomes polar and hence ferroelectric.

We have prepared coherent (010) oriented DyFeO₃ thin films on (010) YAlO₃ substrates to investigate the impact of compressive lattice strain on magnetic properties. For thinner films of around 10 nm, compressive strain is of the order 2-3% and decreases down to < 1% for films with a thickness of 100nm or more. As a consequence of the growth induced strain, the spin reorientation temperature increases to 70 K and above with a transition width of several 10 K. This is unlike for single crystals where the transition width is a few K. Temperature dependent magnetization measurements between 350 K and 2 K are strongly magnetic field dependent and the sign of the slope changes depending on the measurement field, typically between 10 and 100 mT. The sign change requires a two-spin system where the rigid Fe-spin lattice is influencing Dy spins and aligning them antiparallel for small fields and parallel for larger fields. As a consequence, Fe and Dy moments are adding up leading to large magnetic moments for these films of several μ_B . This is evidence of a magnetic symmetry breaking by lowering the magnetic symmetry and therefore giving rise to a polar component. Probing the in-plane electrical polarization using an interdigitated design, we have been able to measure an electrical polarization of $\sim 1 \mu\text{C}/\text{cm}$ along the [100] direction up to 250K, and non along [001] showing that the polar vector P is pointing along [100] unlike for a crystal where P is pointing along [001]. Growth induced compressive strain therefore modifies magnetic and polar properties of epitaxial DyFeO₃ films well beyond what is known for single crystals with a ferroelectric phase close to room temperature.

The quantum metric of the (111) LaAlO₃/SrTiO₃ interface

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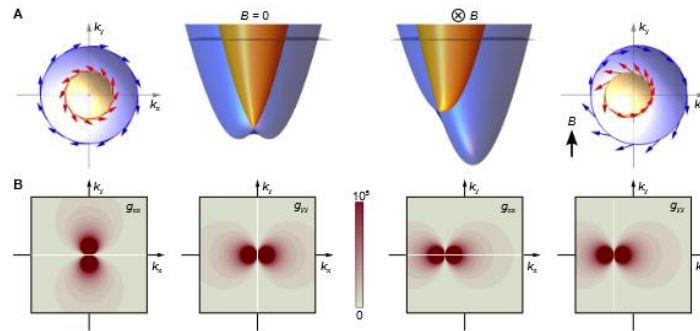
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The electronic responses of quantum materials are governed by the geometry and topology of electronic wavefunctions. These properties are encoded in the Berry curvature and quantum metric, which are respectively the real and imaginary parts of the quantum geometric tensor. The Berry curvature has been probed by measurements of the anomalous Hall effect in ferromagnets and the quantum Hall effect in 2D electron gases [1]. In addition, its higher order components (the Berry curvature dipole and quadrupole) have been associated with nonlinear Hall effects [2-3]. In comparison, the quantum metric has only been observed in topological antiferromagnets and always disjointedly from the Berry curvature [4-5].

Here [6], we theoretically predict and experimentally demonstrate that the spin-momentum locking – a property commonly found at the surfaces and interfaces of materials with spin-orbit coupling – is endowed with a nontrivial quantum metric. This intrinsic feature of spin-locked electronic bands activates a nonlinear in-plane magnetoresistance that we observe and control by voltage gating in the 2D electron gas at the (111)-oriented LaAlO₃/SrTiO₃ interface. The concomitant detection of the quantum metric magnetoresistance and the planar Hall effect driven by the Berry curvature allows us to sense the full quantum geometric tensor in a nonmagnetic material. These findings reveal the importance of the quantum geometry for a vast class of materials, namely, all systems with spin-locked electronic bands, and provide new strategies to design functionalities based on the geometric properties of electronic wavefunctions.



A Spin-locked electronic bands induced by the Rashba effect in the 2D electron gas at the (111)-oriented LaAlO₃/SrTiO₃ interface in the absence (left) and presence (right) of a planar magnetic field. **B** Reciprocal space maps of the diagonal components g_{xx} and g_{yy} of the quantum metric of the Rashba bands at zero and nonzero planar magnetic field.

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Ferromagnetic superconductive two-dimensional electron gases at (110) and (111) oriented epitaxial $\text{KTaO}_3/\text{LaTiO}_3$ interfaces

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Novel functional phenomena can emerge at the interfaces of two distinct materials, with oxide heterostructures serving as excellent platforms for such exotic phenomena, particularly two-dimensional electron gases (2DEGs), superconductivity or ferromagnetism [1,2]. Here, at the interface of the nonmagnetic band insulator KTaO_3 (KTO) and the antiferromagnetic Mott insulator LaTiO_3 (LTO), we observed the evolution of high mobility 2DEGs along the (111) and (110) crystallographic orientations [3]. Interestingly, our 2DEGs exhibit interfacial induced ferromagnetism, as evidenced by electrical and magnetic transport measurements, and supported by x-ray magnetic circular dichroism (XMCD) and density functional theory (DFT) calculations. Remarkably, we observed the coexistence of ferromagnetism and superconductivity at the KTO/LTO interfaces, particularly along the KTO (111) faces. In this work, we discuss the interfacial origin of the ferromagnetism at the KTO/LTO interfaces and explore the correlations between superconductivity and ferromagnetism.

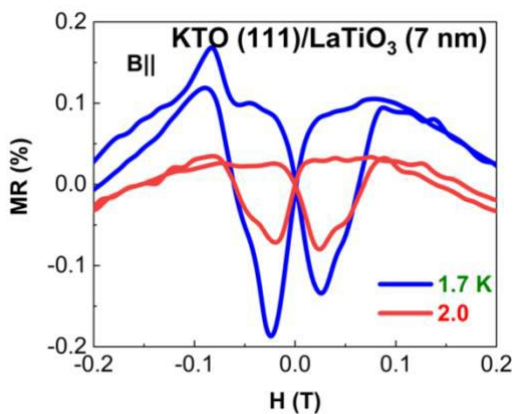


Figure 1. The magnetic field dependence of electrical R_{xx} resistance shows the butterfly-shaped magnetoresistance displaying a hysteresis loop by sweeping the field direction which is direct evidence of the ferromagnetic ordering in the two-dimensional electron gases (2DEGs) the $\text{KTaO}_3/\text{LaTiO}_3$ interface at various temperatures 1.7 K and 2 K (Submitted work, Marzouk et al., [3]).

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Ultrathin Fe on SrTiO₃: Tunable 2D electron and 2D hole states at the interface

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Oxide electronics provide key concepts for enhancing silicon-based semiconductor technologies with novel functionalities. The ability to switch the type of conductivity at SrTiO₃ (STO)-based oxide interfaces between negatively (n) charged electrons or positively (p) charged holes is a missing puzzle piece in this field. While emerging 2DES at oxide-oxide interfaces are a well-studied phenomenon, direct evidence for 2D hole dispersion remains elusive to date – and the respective physical origins of 2DES and 2DHS are still subject to intense debate.

By measuring resonant angle-resolved photoelectron spectroscopy (resARPES), we provide direct evidence for individually emerging n- or p-type 2D band dispersions in STO-based heterostructures [1]. The key parameter to tuning the carrier character of a Fe/STO interface is the oxidation state of the Fe overlayer. For Fe and FeO, we find that hole bands emerge in the empty bandgap region of STO, while for Fe₃O₄ overlayers, a 2D electron system is formed. Using X-ray photoelectron spectroscopy, unexpected oxygen vacancy characteristics arise for the hole-type interfaces, which as of yet had been exclusively assigned to 2D electron gases. To investigate the tunability of the oxygen transfer, we grow high-quality ultrathin Fe films on SrTiO₃ substrates by systematically varying the growth parameters, e.g. (i) growth temperature, (ii) substrate annealing, and (iii) Fe film thickness [2].

This presentation discusses the effect of different growth temperatures, substrate annealing procedures, and film thicknesses and oxidation states of the Fe films on the interfacial properties. The oxidation state of Fe as well as the concentration of defects in SrTiO₃ strongly influences the valence band alignment between electron and hole band bending. This finding opens up the exciting possibility to switch the type of conductivity at STO interfaces by manipulating the oxidation state of the Fe_xO_y overlayer.

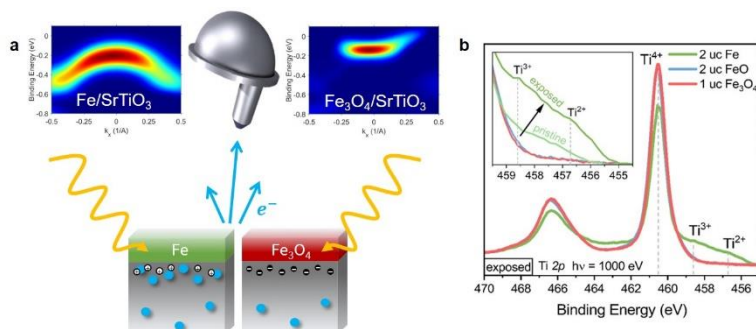


Fig. 1: a) Resonant ARPES measurements revealing the formation of 2D electron and 2D hole states dependent on the respective Fe_xO_y overlayer in STO-based heterostructures. b) Photoemission spectra of the Ti 2p core level at the Fe_xO_y/STO interfaces. Surprisingly, an enhanced Ti^{4+/3+} ratio is observed for the 2D hole dispersion (Fe/STO), which is commonly interpreted as a fingerprint for the emergence of a 2D electron system.

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Direct evidence for inversion symmetry breaking in RE nickelates

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Rare earth nickelate perovskites (RENiO_3) are well-known for their complex phase diagrams, displaying metal-insulator transitions and magnetic ordering that are highly responsive to external factors such as pressure, temperature, and doping. Utilizing polarization-resolved second harmonic generation (SHG), we have detected a second harmonic signal that emerges simultaneously with the metal-to-insulator transition. This observation goes beyond theoretical predictions [1] and experimental evidence [2] of inversion symmetry breaking and the emergence of a static electric dipole as they were always associated with the onset of long-range magnetic ordering.

In this study, we employed a novel experimental technique enabling single-shot measurement of normal and fixed incidence polarization dependence of SHG signals on micron-sized objects. Our optical setup combines a Bessel-Gaussian laser beam with a patented optical module [3], ensuring accurate polarization patterns. We developed a fitting procedure based on evolutionary algorithms, demonstrating high efficiency in reconstructing the second-order nonlinear optical susceptibility tensor, even for materials with the lowest symmetry, thus identifying the most appropriate crystallographic space group.

Applying this method to YNiO_3 indicates that the space group should be $P1$ when the system enters the paramagnetic insulating state, a much lower symmetry than previously accepted for the rare earth nickelates family. Introducing ferroelectricity as an additional degree of freedom to the already diverse physical properties of these compounds is expected to open new avenues for both fundamental research and practical applications.

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***In situ* preparation and ARPES study of superconducting infinite-layer lanthanum nickelates**

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Revealing the momentum-resolved electronic structure of infinite-layer nickelates is essential for understanding this new class of unconventional superconductors, but has been hindered by the formidable challenges in improving the sample quality. In this work, we report for the first time the angle-resolved photoemission spectroscopy of superconducting $\text{La}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films prepared by molecular beam epitaxy and *in situ* atomic-hydrogen reduction [1,2]. The measured Fermi topology closely matches theoretical calculations, showing a large Ni- $d_{x^2-y^2}$ derived Fermi sheet that evolves from hole-like to electron-like along k_z , and a three-dimensional (3D) electron pocket centered at Brillouin zone corner. The Ni- $d_{x^2-y^2}$ derived bands show a mass enhancement (m^*/m_{DFT}) of 2-3, while the 3D electron band shows negligible band renormalization. Moreover, the Ni- $d_{x^2-y^2}$ derived states also display a band dispersion anomaly at higher binding energy, reminiscent of the waterfall feature and kinks observed in cuprates.

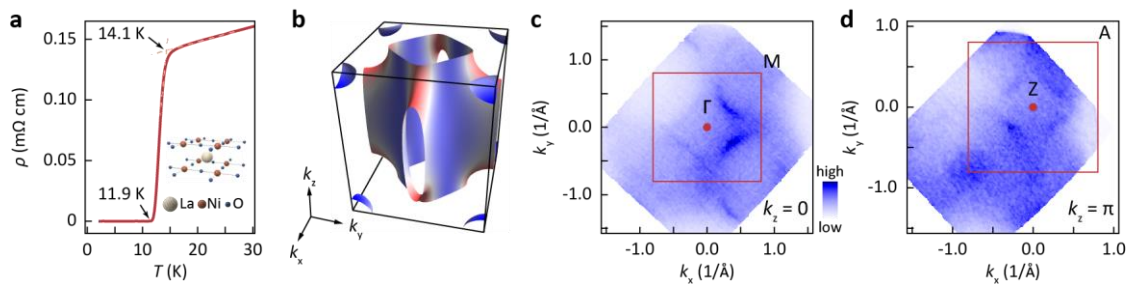


Figure 1. Transport characteristic and Fermi surface topology for superconducting $\text{La}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ film.

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Interface- and defect-induced quantum phases in nickelate thin films and superlattices

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The nickelate family hosts a variety of intriguing phenomena, including magnetic, structural and metal-to-insulator transitions [1]. The recently discovered superconductivity in infinite-layer nickelates [2] has spurred intensive research to unravel its origin. Based on insight from density functional theory calculations with a Hubbard U term, I will focus on the role of the film geometry [3], the recently resolved unexpected structure of the interface [4], as well as the magnetic interactions in the system [5]. Moreover, starting from confined perovskite superlattices, I will address the role of oxygen vacancies giving rise to a stripe antiferromagnetic order [6]. Last but not least, LaNiO_{3-x} is a promising electrocatalyst for the oxygen evolution reaction (OER). To this end, the role of the surface orientation and facet-dependent surface transformation into a NiOOH_x layer on the OER activity will be assessed [7].

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Topotactic reduction effect in infinite-layer nickelates

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Topotactic reduction utilizing metal hydrides as reagents emerges as a highly effective approach to achieve exceptionally low oxidization states of transition metal ions and create unconventional coordination networks. This method opens avenues to the development of entirely new functional material families that boast novel physical properties. A notable example is the discovery of superconductivity in nickelates, in which the perovskite precursor undergoes a transformation into an infinite-layer structure. However, the reduction effect on the atomic reconstruction and electronic structures, which is crucial to the emergence of superconductivity in nickelates, remains largely unresolved. Here we design two sets of $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films, with pure infinite-layer phase and mixed phases respectively, to address the origin of reduction effect on the superconductivity. Secondary ion mass spectroscopy shows negligible hydrogen intercalation in pure infinite-layer phase, highlighting that hydrogen is not necessary for superconductivity. X-ray absorption spectroscopy shows a significant linear dichroism with dominant Ni $3d_{x^2-y^2}$ orbitals on superconducting samples, indicating a Ni single-band nature of infinite-layer nickelates. Superconductivity is found to manifest dome-like reduction duration dependence, consistent with the behaviour of Ni $3d$ orbitals asymmetry. Our results unveil the critical role of reduction in modulating the Ni- $3d$ orbital polarization and its impact on the superconducting properties [1].

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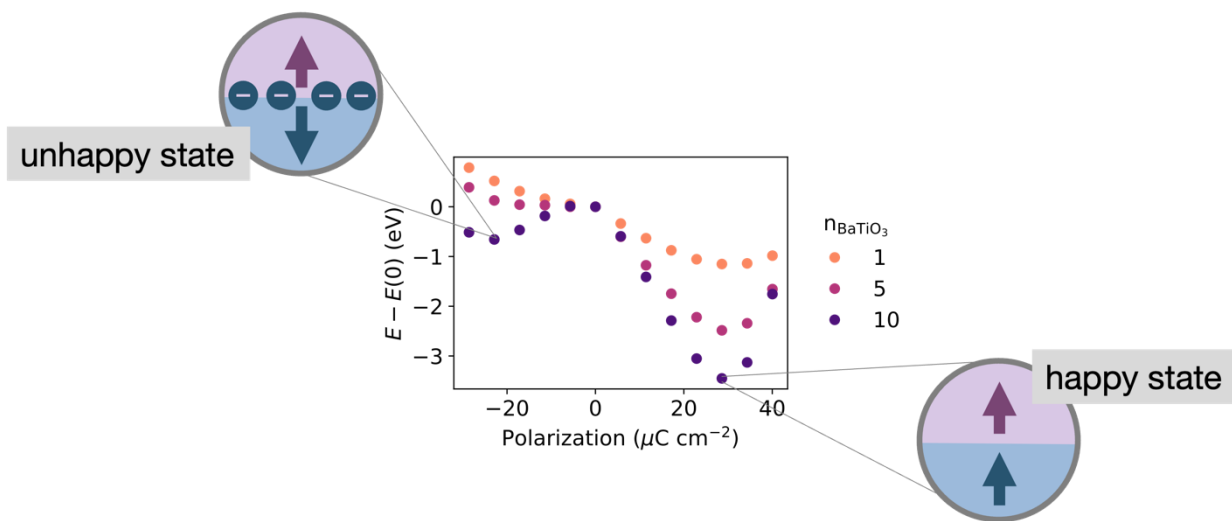
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Interplay of metallicity, ferroelectricity and layer charges in SmNiO_3 and BaTiO_3 superlattices

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We present a study of the effect of the polar discontinuity in superlattices containing metallic SmNiO_3 (SNO) and ferroelectric BaTiO_3 (BTO) using density-functional theory (DFT) and an electrostatic model. The interface of SNO and BTO hosts various interesting functionalities: Since BTO and SNO are II-IV and III-III perovskites respectively, the different layer charges lead to a built-in polar discontinuity in addition to that arising from the spontaneous polarization of BTO. These two sources of polarization, in combination with the metallic screening of SNO, have a complex interplay. Using DFT, we find that, despite the metallicity of SNO, the layer polarization of the metal strongly affects the ferroelectric ground state. The system avoids a polar discontinuity by aligning the spontaneous polarization of the BTO parallel to the layer polarization of the SNO in the so-called ‘happy’ orientation. In the ‘happy’ orientation, BTO shows a vanishing critical thickness, in contrast to the 6-unit-cell critical thickness previously found for BTO with II-IV metallic electrodes [1]. However, the opposite ‘unhappy’ polarization orientation is highly disfavoured for thin films, with the stability depending strongly on the thickness of the BTO layer and the nature of the tilts within the SNO layer. We describe the behaviour of the superlattices using a simple electrostatic model and find the metastability of the ‘unhappy’ state to increase with the layer thickness. We conclude that layer charges of a metal in ferroelectric capacitors cannot be neglected and in fact could provide new ways to control the ferroelectric properties.



Energy profile for the polarization of BTO in BTO-SNO superlattices. The ‘happy’ state, i.e. the polarization of BTO (blue arrow) aligning with the layer polarization of SNO (purple arrow) is energetically favored compared to the opposite ‘unhappy’ orientation.

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Engineering unequal antipolar displacement in ferromagnetic layered oxide heterostructures

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Multiferroic materials that simultaneously display ferroelectricity and magnetic ordering are highly sought after for next-generation electronics applications. The combination of these properties is very rare in single-phase materials, thus heterostructure engineering represents a promising alternative pathway. In this context, heterostructures comprising the insulating and ferromagnetic double perovskites $\text{La}_2\text{NiMnO}_6$ and $\text{RE}_2\text{NiMnO}_6$ (RE=rare earth \neq La) represent an intriguing system. These superlattices are predicted to exhibit unequal antipolar displacement of the La and RE ions [1], which, when combined with odd periodicity superlattice layering, could potentially lead to hybrid improper ferroelectricity.

$\text{La}_2\text{NiMnO}_6/\text{Sm}_2\text{NiMnO}_6$ superlattices are grown with atomic precision using a sputtering system equipped with high-energy electron diffraction (RHEED). The heterostructures display robust ferromagnetism [2], as confirmed by in-house magnetometry and synchrotron measurements. Scanning transmission electron microscopy (STEM) in conjunction with first-principles calculations has validated the predicted unequal antipolar displacement in our superlattices. This represents a significant advance towards the establishment of hybrid improper ferroelectricity in artificially layered heterostructures.

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Mapping the complex evolution of ferroelastic/ferroelectric domain patterns in epitaxially strained PbTiO₃ heterostructures

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In ferroelectric thin films, the complex interplay between mechanical and electrostatic boundary conditions allows for the formation of a large variety of domain structures with fascinating properties. These domain structures not only change the properties of the ferroelectric itself, but can also be used to change the properties of other materials through electrostatic and structural coupling.

In this work, we study the complex ferroelastic/ferroelectric domain structure in the prototypical ferroelectric PbTiO₃. We investigate epitaxially strained structures grown on (110)_o-oriented DyScO₃ substrates, using a combination of atomic force microscopy, laboratory and synchrotron x-ray diffraction and high resolution scanning transmission electron microscopy. We observe that the anisotropic strain imposed by the orthorhombic substrate creates a large asymmetry in the domain configuration, with domain walls macroscopically aligned along one of the two in-plane directions. We show that the periodicity as a function of film thickness deviates from the Kittel law. As the ferroelectric film thickness increases, we find that the domain configuration evolves from flux-closure to *a/c*-phase, with a larger scale arrangement of domains into superdomains.

Above a certain critical thickness, the large structural distortions associated with the ferroelastic domains propagate through the top SrRuO₃ layer, creating a modulated structure that extends beyond the ferroelectric layer thickness, leading to nanoscale domain engineering in SrRuO₃ thin films.

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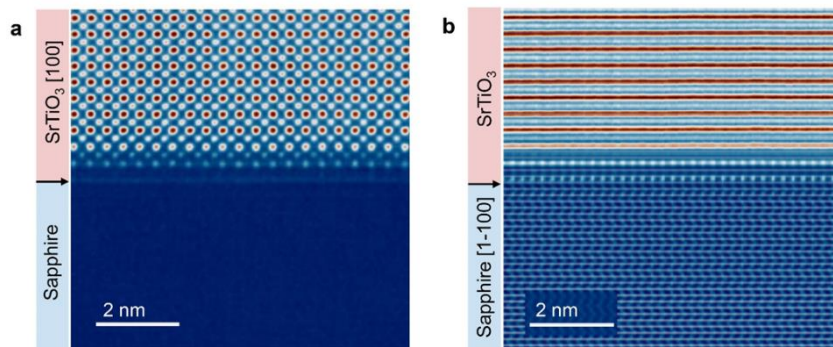
Mapping the complex evolution of ferroelastic/ferroelectric domain patterns in epitaxially strained PbTiO₃ heterostructures. Lichtensteiger, Hadjimichael, Zatterin, Su, Gaponenko, Tovaglieri, Paruch, Gloter, & Triscone (2023). APL Materials, 11(061126). [[link](#)]
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Interface design beyond epitaxy

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The study of thin films, their interfaces and thin film heterostructures has been a cornerstone of experimental research at reduced dimensions. Advancements in epitaxial techniques have enabled the engineering of atomically precise and well defined interfaces. However, the parameter space of interfaces enabled by epitaxy is determined by the lattice symmetry of the substrate and the plane on which epitaxy takes place. This for such interfaces is taken as a given. I will take this opportunity to focus on recent developments in oxide growth enabling the separation of the grown thin films from the growth substrate, resulting in freestanding oxide membranes [1]. These membranes have allowed for unprecedented access to avenues in oxides, with novel symmetry disallowed interfaces, which go beyond the epitaxially possible atomically sharp interfaces [2]. I will show striking phenomena we have observed with an exemplary SrTiO₃ (001) on a sapphire (0001) interface, through which I hope to convey how these developments in oxides promise a fertile ground for remarkable discoveries in material science and physics.



HAADF-STEM images of cross sections of SrTiO₃-sapphire heterojunctions viewed along (a) SrTiO₃ [100] and (b) sapphire [1-100]. The arrows mark the nominal interface between the sapphire and the SrTiO₃.

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Reconfiguring topology in free-standing antiferromagnetic membranes

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Antiferromagnets hosting real-space topological textures are promising platforms to model fundamental ultrafast phenomena and explore spintronics [1,2]. However, they have only been fabricated epitaxially on specific symmetry-matched substrates, thereby preserving their intrinsic magneto-crystalline order [3,4,5]. This curtails their integration with dissimilar supports, restricting the scope of fundamental and applied investigations.

I will discuss how we circumvented this limitation by designing detachable crystalline antiferromagnetic nanomembranes of α -Fe₂O₃ [6]. First, using transmission-based antiferromagnetic vector mapping, we showed that flat nanomembranes host a spin reorientation transition and rich topological phenomenology. Second, we exploited their extreme flexibility to demonstrate the reconfiguration of antiferromagnetic states across 3D membrane folds resulting from flexure-induced strains. Finally, we combined these advances using a controlled manipulator to realise strain-driven non-thermal generation of topological textures at room temperature. Integration of such free-standing antiferromagnetic layers with 3D nanostructures could enable spin texture designs via magnetoelastic-/geometric-effects in the quasi-static and dynamical regimes, opening new explorations into curvilinear antiferromagnetism and unconventional computing.

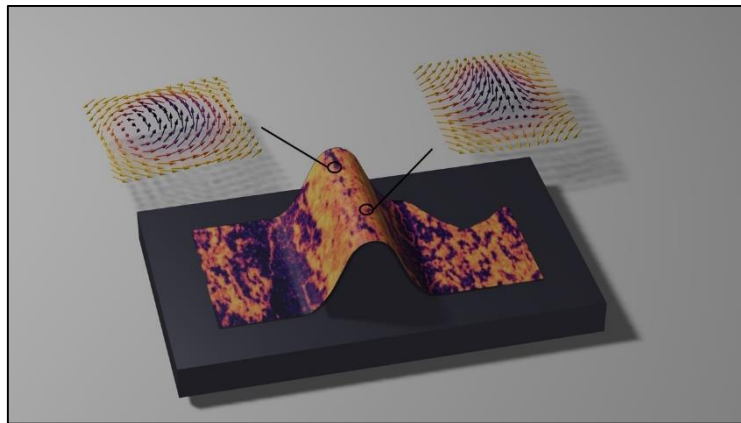


Figure 1: Flexure-driven spatial reconfiguration of antiferromagnetic states and stabilization of topological solitons – (anti)merons and bimerons (insets) – in an α -Fe₂O₃ membrane at room temperature [6].

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Defect thermodynamics and engineering in transferable oxide thin films and bilayer heterostructures

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Free-standing oxides based on the delamination of atomically defined epitaxial thin films provide new opportunities to combine functional complex oxides with semiconductor (silicon) electronics. At the same time, the nanoscale confinement of these transferable lamellae enables to synthesize unique defect structures and functionalities, that follow from the structural boundary conditions and limited dimensions. Here we discuss the synthesis of transferable perovskite oxide lamellae via the all-perovskite sacrificial-layer route as compared to the graphene-buffered remote epitaxy approach via PLD [1]. We then explore the defect structure of transferred lamellae based on the example of SrTiO₃, serving as a model system for ionic-electronic phenomena. As we elaborate, the confinement of the lamella facilitates the overall formation of oxygen vacancies as compared to the bulk oxide. Exploiting the ion-transfer between SrTiO₃ and LaAlO₃ (deposited under low oxygen pressure), we address the local redox-activity and demonstrate defect concentrations beyond the thermodynamic limit in the transferred lamella [2].

Finally, we demonstrate how the transferred SrTiO₃ lamellae can be used as a template for the growth of functional bilayer structures hosting ion-driven magnetic-electronic phase transitions and electrocatalytic properties. Achieving a layer-by-layer growth mode, we show that bilayer structures with sufficiently defined interfaces and morphology can be obtained on a non-epitaxial substrate, enabling a detailed analysis of magnetic and chemical depth profiles. Notably, despite the lack of an epitaxial relationship between substrate and bilayer, the lateral strain state of the bilayers depends on the expected bond-type (ionic vs. covalent) between the as-transferred lamella and substrate, allowing to achieve novel strain states that are inaccessible in standard epitaxy. These examples showcase that the integration of complex transferable oxides heterostructures with semiconductors will come with new opportunities but also new challenges to tailor and design the properties of functional oxides in their new environment.

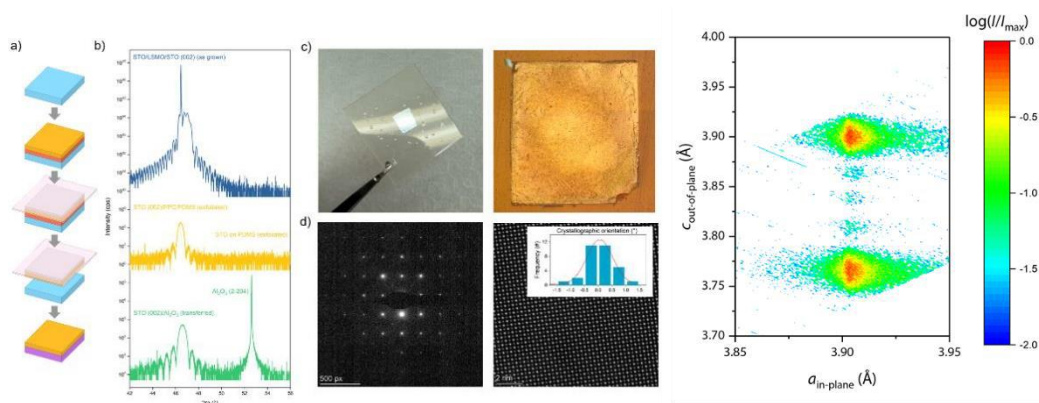


Fig. 1: Lamella fabrication and structural relationships in La,SrCoO₃/SrTiO₃ bilayer heterostructures.

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Emerging phase transitions in lead-free antiferroelectric heterostructures and membranes

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As one of the prominent lead-free antiferroelectric alternatives, sodium niobate (NaNbO_3) has garnered considerable research attention due to its significant potential in energy storage applications. Known for being one of the most structurally complex perovskite materials, NaNbO_3 exhibits a diverse set of structural phases characterized by distinct symmetries, ferroic orders, and oxygen octahedral tilting patterns. This intricate energy landscape in NaNbO_3 opens up possibilities for inducing and controlling novel phase transitions via strain, thickness, and electric field.

In this presentation, I will introduce our recent studies on the intrinsic size dependence of antiferroelectricity in freestanding NaNbO_3 membranes by decoupling the substrate effects. Through a wide range of experimental and theoretical approaches, we have probed an intriguing surface-driven antiferroelectric-to-ferroelectric transition that occurs as the membrane thickness decreases. Additionally, I will present our recent findings regarding the strain-induced phase transition in NaNbO_3 heterostructures. We observed an emerging mixed phase state with coexisting monoclinic and orthorhombic phases in NaNbO_3 , leading to large field-driven effects. Our work demonstrates the enormous potential of using NaNbO_3 as a fertile ground for exploring emerging phase transitions and phenomena within the family of complex oxides.

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Ferroelectricity, chirality, spin-orbit coupling and cavity states in $\text{Pb}_5\text{Ge}_3\text{O}_{11}$ crystal

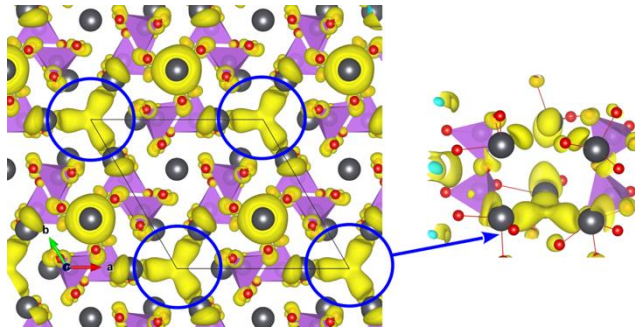
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$\text{Pb}_5\text{Ge}_3\text{O}_{11}$ is a room temperature ferroelectric, which is also known to be gyroelectric, i.e. its natural optical activity can be reversed under an applied electric field during the ferroelectric domain switching. It is recently re-explored because of his unique topological and antiferroelectric domains coming from the uniaxial ferroelectricity and head-to-head/tail-to-tail domain walls [1,2,3]. Here, we will show that $\text{Pb}_5\text{Ge}_3\text{O}_{11}$ is even more versatile than known so far. By scrutinizing this crystal with first-principles DFT calculations, we will first show that the ferroelectric character belongs to the so-called hyperferroelectrics where the polar distortions uniquely couple non-linearly and cooperatively with the internal degrees of freedom of the paraelectric phase (i.e. the invariant modes). We will also quantify the origin of the structural chirality of the low symmetry ferroelectric phase, which will allow us to explain why the chirality can be flipped by an electric field and, hence, why the crystal is gyroelectric [4]. At last, we will show -that the ferroelectric energy landscape is surprisingly and strongly renormalized by the spin-orbit coupling. In trying to understand why the spin-orbit coupling is so important in this non-magnetic crystal, we will highlight that it is coming from the empty 6p orbitals of the Pb atoms that originally localize in the natural empty tubular cavity presents in this crystal (see Figure). This Pb-6p cavity state exhibits a sizable spin splitting with a mixed Rashba-Weyl character and a topologically protected crossing of the related bands. At last, the cavity state also makes the ferroelectricity very robust against electron doping [5].



Schematic picture of the partial charge density (in yellow) associated with the first conduction band with a strong Pb-6p character. Lead and oxygen atoms are shown in black and red respectively. GeO_4 tetrahedra are shown in purple, and empty channels are evidenced by blue circles. A zoom on the cavity state is shown on the right panel.

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Ultrathin crystalline membranes of ferroelectric hafnia

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Freestanding oxide membranes, which can be transferred onto and make interfaces with any materials, hold great promise for scientific research and device development. It has been shown that freestanding membranes of oxides with bulk-equivalent crystal structures and physical properties can be fabricated. However, freestanding membranes of oxides in metastable phases have yet to be thoroughly explored. In this presentation, we demonstrate that metastable and ferroelectric hafnia ($\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ (HZO)) can be fabricated into the membrane form even down to 1-nm-thick [1].

We fabricated freestanding hafnia membranes from 1- to 5-nm-thick by selectively etching the (La,Sr)MnO₃ (LSMO) buffer layer and exfoliating HZO layer from epitaxial HZO/LSMO/SrTiO₃ heterostructures. We then transferred hafnia membranes to foreign substrates, including TEM grids and Si/SiO₂ wafers, to characterize their crystal structures and electrical properties, respectively. The top-view HAADF-STEM observations in Fig.1(a) reveal the thickness-dependent changes in the crystal structures of HZO membranes. The 1-nm-thick HZO membrane has the metastable rhombohedral structure, which evolves into the metastable orthorhombic structure when thickening up to 5 nm. The positive-up-negative-down (PUND) measurements (Fig. 1(b)) show that regardless of the thickness, the membranes have ferroelectric polarizations of around $13\mu\text{C}/\text{cm}^2$. Our results clarify that hafnia membranes can sustain their metastable crystal structures even down to 1-nm-thick without external support from epitaxial strain, demonstrating the scale-free nature of ferroelectricity in hafnia.

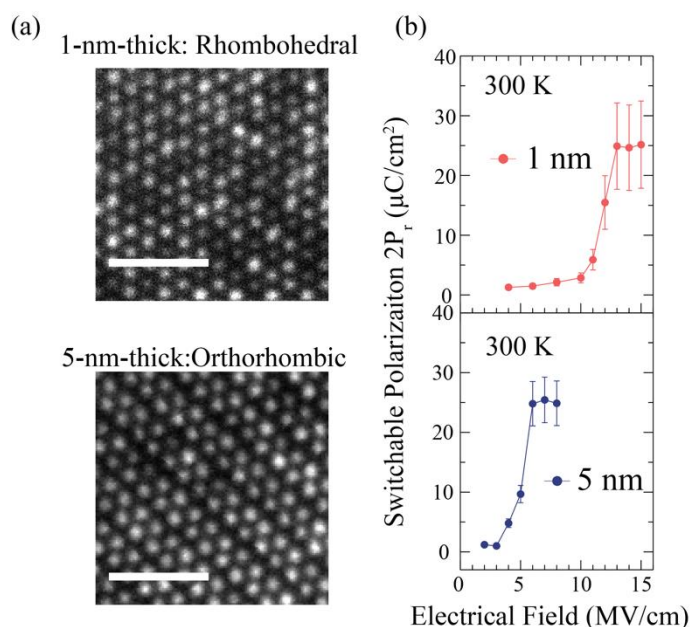


Fig. 1 (a) Top-view HAADF-STEM images and (b) PUND results of 1-nm- and 5-nm-thick hafnia membranes. The scale bar denotes 1 nm.

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Electric field-manipulated optical chirality in ferroelectric vortex domains

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Manipulating optical chirality via electric fields has garnered considerable attention in the realm of both fundamental physics and practical applications. Chiral ferroelectrics, characterized by their inherent optical chirality and switchable spontaneous polarization, have been emerging as a promising platform for electronic-photonics integrated circuits applications. Despite notable advancements in the realm of chirality within organics, the pace of development in inorganic ferroelectrics has been comparatively sluggish, primarily attributed to the scarcity of chiral centers in these materials. Recently, by introducing polar topological textures relying on the discontinuous interface into heterostructures [1, 2], chirality has been successfully achieved within inorganics. However, the generalizability of this approach, which is contingent on the discontinuity of the interface, has yet to be explored across different material systems, and the chirality stemming from polar topological textures is challenging to detect, highlighting the necessity for further investigation in this area.

Expanding upon our previous research [3], we propose an innovative protocol for stabilizing ferroelectric vortex domains in BiFeO₃-based (BFO) nanoislands, and report the observation of optical chirality in spontaneously stabilized vortex domains within a single-phase ferroelectric material, rather than a heterostructure. By integrating the piezoresponse force microscopy and nonlinear optical second-harmonic generation probes, the correlation between chirality and ferroelectric vortex domains in these BFO nanoislands is explicitly established. Capitalizing on the synergistic coupling between ferroelectricity and chirality, we are able to achieve a reversible and nonvolatile electric field-controllable chiral transformation. The self-assembled BFO nanoislands, serving as discrete chiral units, are integrated into ordered arrays and exhibit capabilities in chiral optical display functionalities. This achievement represents a substantial step forward in chirality manipulation within simple inorganic material systems through electrical means, which could have potential implications for the development of advanced optoelectronic devices.

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Electrochemical oxygen intercalation for precise tuning of functionalities in next-generation oxide-based devices

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Doping represents the most effective strategy for achieving modulation of functional properties in oxide systems. However, while cationic doping (substitution) can be easily controlled during material synthesis, tuning of anionic doping (via control of oxygen non-stoichiometry) remains challenging, as traditional techniques (i.e. high temperature annealings) fail to provide modularity. Electrochemical pumping of oxygen represents a superior alternative, in which a precise control of the oxygen content is achieved through voltage-driven (de)intercalation of oxygen ions through a solid electrolyte in a robust, fast, and reversible manner, providing accesses to potentially infinite intermediate states [1].

In the present work, we provide clear examples of how electrochemical oxygen intercalation via application of a Nernstian voltage can be employed for the fundamental investigation of complex oxide systems, as well as for the design of next-generation energy devices. We investigate mixed ionic-electronic conducting (La,Sr)FeO_{3-δ} thin films and we show that potentially infinite intermediate states, characterized by different electronic conductivity spanning over 5 orders of magnitude, can be stabilized by changing the oxygen content in a continuous and controlled way. A smart experimental design allowed for the simultaneous characterization of structural, optical, electrical, and magnetic properties of the films, shedding light on the mechanisms which correlate such properties with the oxygen content and temperature. Furthermore, we demonstrate that this straightforward electrochemical system presents enormous potential for next-generation devices based on oxide ion intercalation, including energy storage and memory devices [2].

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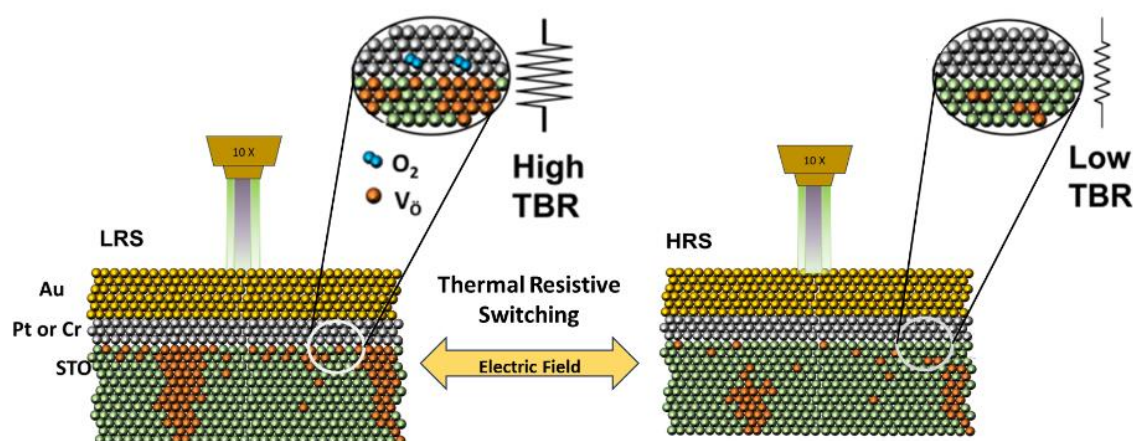
Coupled resistive and thermal switching in Metal/SrTiO₃ devices

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Achieving a dynamic control of the heat flow in crystalline solids is key due to the relentless miniaturization of electronic devices. In this regard, the possibility to modulate the local concentration of oxygen vacancies in transition-metal oxides, associated to the resistive switching effect, has emerged as a possibility of designing thermal switches. In this work, we demonstrate a reversible effect on the thermal boundary resistance across SrTiO₃/Pt, Cr interfaces, associated to an electric field-driven accumulation/depletion of oxygen vacancies [1].



Schematic representation of Metal/SrTiO₃ devices.

Using frequency domain thermoreflectance (FDTR) we measured a variation of 20% in the thermal boundary resistance of Pt/STO and Cr/STO devices, at room temperature. Relaxation experiments demonstrate the correlation between the thermal and electrical resistance states.

Our experiments demonstrate the possibility to modulate the oxygen vacancies at metal-oxide interfaces for thermal switching devices.

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Y₂O₃-based memristive devices

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Oxide-based memristors offer highly tailorable functionalities for memory, storage and neuromorphic applications. In OxRAM devices the material aspects of the switching medium have a very high impact on the performance, and thus material engineering can be utilized to address key challenges in their reliability. Within the family of transition metal oxides yttrium oxide offers good stability, has only a single metastable suboxide and, arising from the cubic bixbyite structure, has an intrinsically high anion vacancy concentration. The influence of these qualities on the resistive switching performance of yttria based memristors manifests as excellent retention characteristics also under heat accelerated aging [1] and high stability at elevated temperatures [2]. We have shown that through oxygen engineering the cubic phase of yttrium oxide can be isolated, achieving analog switching behaviour under both the set and reset processes [3]. While the intrinsic oxygen vacancy network facilitates lower energy barriers for filament formation, resulting in lower switching voltages and power consumption, decreasing the oxygen content also promotes controlled, electric-field-driven switching, resulting in a less robust filament and the appearance of intermediate states that show nonlinear conductance quantization [4]. Furthermore, our most recent results imply that yttria-based devices have superior temporal stability under $>10^{12}$ readouts, even at elevated temperatures in all resistive states, a significant achievement for device reliability.

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Nonlinear and nonreciprocal transport effects in untwinned thin films of topological Weyl metal SrRuO₃

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Youngjoon Han,⁴ Xinwei Li,⁴ Cheng-Tung Cheng,¹ Song Yang,⁵ Chun-Yen Lin,⁵
I-Chun Cheng,⁸ Chia-Hung Hsu,⁵ D. Hsieh,⁴ Wei-Cheng Lee,³ Guang-Yu Guo,^{2,7} and Wei-Li Lee¹

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In topological Dirac and Weyl semimetals, nontrivial conical bands with Fermi-arc surface states give rise to negative longitudinal magnetoresistance due to chiral anomaly effect and unusual thickness dependent quantum oscillation from Weyl-orbit effect, which were demonstrated recently in ferromagnetic Weyl metal SrRuO₃ (SRO) thin films [1]. In this work, we report the experimental observations of large nonlinear and nonreciprocal transport effects for both longitudinal and transverse channels in an untwinned Weyl metal of SRO thin film grown on a SrTiO₃ substrate. From rigorous measurements with bias current applied along various directions with respect to the crystalline principal axes, the magnitude of nonlinear Hall signals from the transverse channel exhibits a simple $\sin\alpha$ dependence at low temperatures, where α is the angle between bias current direction and orthorhombic [001]_o, reaching a maximum when current is along orthorhombic [1-10]_o. On the contrary, the magnitude of nonlinear and nonreciprocal signals in the longitudinal channel attains a maximum for bias current along [001]_o, and it vanishes for bias current along [1-10]_o. The observed α -dependent nonlinear and nonreciprocal signals in longitudinal and transverse channels reveal a magnetic Weyl phase with an effective Berry curvature dipole along [1-10]_o, accompanied by 1D chiral edge modes along [001]_o [2].

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Light-induced metastability in a cuprate ladder

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Intense ultrashort lasers are an extremely effective tool for controlling the properties of quantum materials and inducing emergent states with novel functionalities. Some of the most spectacular light-induced phenomena, such as superconducting-like phases, transient charge density wave ordering, and excitonic condensation, are found to occur in materials dominated by strong electronic correlations with a large susceptibility to external stimuli. A key priority is to stabilize these effects to timescales far beyond the duration of the external excitation with the goal to better understand their microscopic physics and provide a path towards their functionalization. Here, we focus on the problem of electronic metastability in Mott insulators, where prethermal and metastable electronic distributions are necessary for the emergence of novel phenomena such as η -pairing condensation and light-driven superconductivity [1]. By studying a paradigmatic system, the cuprate ladder oxide $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$, we demonstrate the presence of a metastable electronic charge separation enabled by a transient symmetry breaking phenomenon [2]. Ultrafast x-ray spectroscopy [3,4,5] enables interrogating the microscopic charge and spin dynamics of the metastable phase with unprecedented detail, and quantifying the intra-unit-cell charge transfer. Our findings open a new route towards *on-demand* control of charge separation in layered systems coupled to charge reservoirs.

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Néel skyrmion bubbles in $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.95}\text{Ru}_{0.05}\text{O}_3$ multilayers

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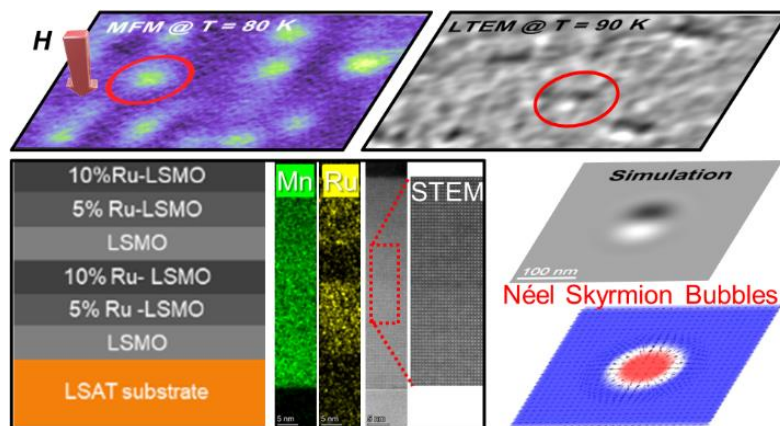
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Ferromagnetic $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ru}_x\text{O}_3$ epitaxial multilayers with controlled variation of the Ru/Mn content were synthesized to engineer canted magnetic anisotropy and variable exchange interactions, and to explore the possibility of generating Dzyaloshinskii–Moriya interaction. The ultimate aim of the multilayer design is to provide the conditions for the formation of domains with non-trivial magnetic topology in an oxide thin film system. Employing magnetic force microscopy and Lorentz transmission electron microscopy in varying perpendicular magnetic fields, magnetic stripe domains separated by Néel-type domain walls as well as Néel skyrmions smaller than 100 nm in diameter were observed [1]. These findings are consistent with micromagnetic modelling taking into account a sizeable Dzyaloshinskii–Moriya interaction arising from the inversion symmetry breaking and possibly from strain effects in the multilayer system.



Magnetic force microscopy and Lorentz transmission electron microscopy enabled the study of bubble domains in $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ru}_x\text{O}_3$ multilayers and their texture is indicative of a Néel bubble structure.

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Integration of high- T_c superconductors and high- Q -factor mechanical resonators in LaAlO_3 suspended micro-bridges

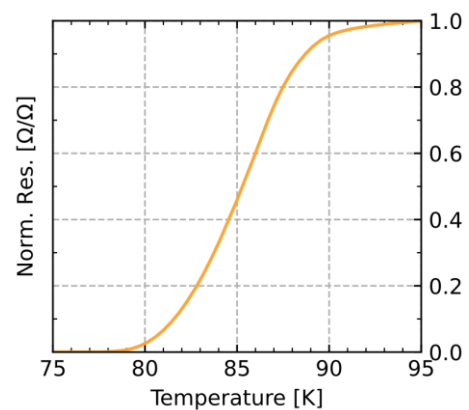
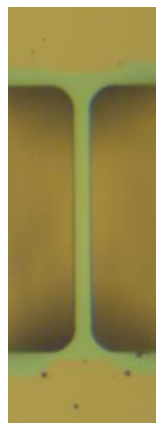
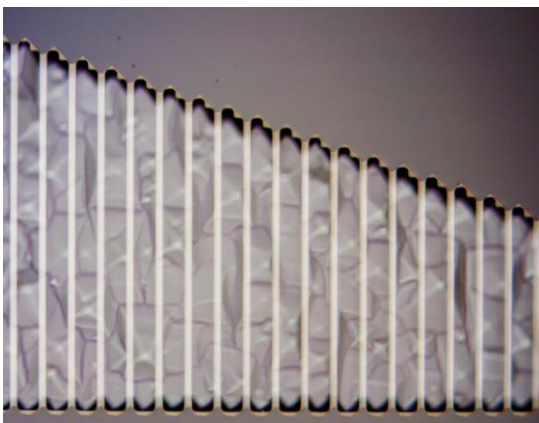
Nicola Manca¹, Alexei Kalaboukhov², Alejandro E. Plaza¹, Leonélio Cichetto Jr¹, Eric Walhberg², Emilio Bellingeri¹, Francesco Bisio¹, Floriana Lombardi², Daniele Marré^{3,1}, and Luca Pellegrino¹

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Micro-mechanical resonators are building blocks of a variety of applications in basic science and applied electronics. This device technology relies on well-established and reproducible silicon-based fabrication processes with outstanding performances in term of mechanical Q -factor and sensitivity to external perturbations. Broadening the functionalities of MEMS is a key step for both applied and fundamental science, but combining functional materials and silicon-based compounds is challenging. An alternative route to face this issue is offered by complex oxides, as they offer the opportunity to grow heterostructures made of materials inherently showing a variety of physical properties. As a demonstrator of the potential of this approach we report on the integration of $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO), a high- T_c superconductor, with high Q -factor micro-bridge resonators made of a single-crystal LaAlO_3 (LAO) thin film. LAO resonators are highly tensile-strained, show Q -factor in the range of hundred of thousands, and have low roughness, making them ideal candidates for the integration of other oxides by ex-situ growth. YBCO was deposited by pulse laser deposition on top of LAO micro-bridges and YBCO/LAO bridges show superconducting transition with zero resistance below 78 K [1]. This result opens new possibilities towards the development of advanced micro-mechanical transducers, such as bolometers or magnetic field detectors, as well as basic science experiments in solid state physics, material science, and quantum opto-mechanics.



(left) harp of LAO microbridge resonators; (center) a LAO microbridge after the deposition of 100 nm of YBCO; (right) normalized electrical resistance of the LAO/YBCO microbridge as a function of temperature.

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VO₂ driven electromechanically reconfigurable THz devices

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Dynamic terahertz devices are essential for the development of the next generation of wireless communication, sensing and non-destructive imaging technologies. Metasurfaces have emerged as paradigm-shifting platform, offering diverse functionalities, miniaturization, and simplified fabrication compared to their 3D counterparts. However, in-plane mirror symmetry and limited degrees of freedom restrict advanced chiral responses, beamforming, and reconfiguration capabilities. Dynamic tunability of these metasurfaces is critical and has been a focus of research for decades. Among various approaches, integrating VO₂ has been key for achieving tunability in planar THz metasurfaces due to the strong contrast in conductivity and permittivity across the insulator-to-metal transition at 67°C [1]. However, simultaneous changes in conductivity and permittivity can pose challenges, limiting effectiveness in some applications [2]. In this work, we demonstrate a robust and scalable platform for multifunctional terahertz metadevices, featuring electrically actuated resonators that can be colossally reconfigured between planar and 3D geometries using the phase transition in VO₂ [3]. The tunable response arises from changes in the resonator structure, not the material properties of VO₂. To illustrate this platform, we fabricated metadevices with 3D Split Ring Resonators, incorporating two counteracting forces: i) folding induced by stress mismatch, enabling non-volatile state design, and ii) unfolding triggered by the strain during the insulator-to-metal transition in VO₂, allowing volatile structural reconfiguration. This extensive structural reconfiguration space permits resonance mode switching, highly tunable magnetic and electric polarizabilities, and enhanced frequency agility. Moreover, the hysteretic phase transition of VO₂ enables multi-state memory. Therefore, these VO₂ integrated metadevices are electrically reconfigurable, multifunctional, spectrally scalable, and technologically adaptable, making them highly suitable for 6G communication devices such as reconfigurable intelligent surfaces, holographic beam formers, and spatial light modulators.

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Ion irradiation induced surface electronic structure variation in the epitaxial lanthanum nickelate based heterostructures

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Lanthanum nickelate (LaNiO₃; LNO) is a conducting oxide which widely utilized as electrode material owing to their ease of integration with epitaxial functional oxides utilized in ferroelectric, resistive switching materials [1,2], etc. The surface electronic structure of LNO undergoes various changes under different external environmental conditions like ambient air, temperature, etc [1,2], and have potential effect on the surface electrical transport properties, e.g. metal-work function, pinning of Fermi level, and density of states (DOS). Swift heavy ion irradiation is a well-known tool to modify the properties of material by utilizing appropriate ion-type and energy [3]. In this work [4], an effort is focused to understand the changes in the surface electronic structure of the epitaxial LNO thin films produced by swift heavy ion irradiation (Ag¹⁵⁺(200 MeV), utilizing a surface-sensitive La (4d to 4f) and Ni (3p to 3d) photoemission transitions in the pristine, highest (10¹² ions/cm²), medium (5 × 10¹¹ ions/cm²), and lowest (10¹¹ ions/cm²) ion-doses of irradiation using angle integrated photoemission spectroscopy (AIPES). The Super-Coster Kronig induced slight enhancement in the spectral features around ~115–116 eV photon energy for La (4d to 4f) transitions were observed, with absence of such enhancement for Ni (3p to 3d) transitions in the pristine and irradiated samples. Moreover, an alteration in the constant initial state (CIS) spectra for La (4d to 4f) transitions and presence of a surface states close to the Fermi level in the valence band spectra for Ni (3p to 3d) transitions, with a different ion-doses found to be associated with the oxygen vacancies. Interestingly for Ni (3p to 3d) transition spectra, the density of states (DOS) at the Fermi Level was found to increase orderly with the irradiation dose, and a systematic correlation between the DOS at Fermi level and the surface states was found in the pristine and irradiated samples. For the highest dose sample, the surface states lie above the Fermi level has highest DOS at the Fermi level, which is due to upward shift of the surface states towards the Fermi level with an increase in the irradiation dose.

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Superconductivity and quantum criticality in two-dimensional NbO₂ layers

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[Introduction] In cuprate high- T_c superconductors, superconductivity occurs in the two-dimensional CuO₂ layers. The parent compound is an antiferromagnetic (AF) Mott insulator with a d^9 electron configuration, and the Fermi level is located in a half-filled single band, $3d_{x^2-y^2}$ orbital, realizing a pronounced strong electron correlation. Carrier doping to such AF phases suppresses the magnetic interaction, resulting in the appearance of superconducting domes [1]. However, the relationship between the magnetic interaction and superconductivity remains an open question. The number of compounds that exhibit such "strongly correlated superconductivity" is very limited to heavy-fermion systems, iron-based compounds, and Moiré superlattices [2]. In this study, we focused on superconducting Li_{1-x}NbO₂, which have two-dimensional $2H$ -type NbO₂ layers and a correlated single-band electronic structure [3]. To obtain the electronic phase diagram, an electrochemical cell with Li_{1-x}NbO₂ epitaxial films as an electrode was fabricated [4], and transport properties were measured while precisely controlling the carrier concentration in the NbO₂ layer.

[Results and Discussion] The host LiNbO₂ showed insulating behavior, and thus it was considered to be a band insulator with a completely filled single Nb $4d_{z^2}$ band; upon deintercalation of Li⁺-ions corresponding to hole doping into the band, Li_{1-x}NbO₂ showed a transition to metal and eventually, a superconducting dome (T_c) appeared (Fig. 1) at larger hole carrier concentrations. In addition, a non-Fermi-liquid (FL) behavior was observed in which the temperature dependence of the resistivity shifted from T^2 - to T -linear relationship. The transition temperature to the FL states (T_{FL}) decreased linearly with hole carrier concentration. These results suggest that two-dimensional AF fluctuations are enhanced near the superconducting dome, which is known as a strange metallic behavior. As a result, the electronic phase diagram of Li_{1-x}NbO₂ has features very similar to those of exotic superconductors, in which magnetic interactions mediate the formation of Cooper pairs. Therefore, two-dimensional NbO₂ layers would be a new platform for investigating strongly correlated superconductivity. The present study has also revealed physical properties that may be unique to the frustrated magnetisms in the triangular lattice, such as the observation of the Kondo effect (T_{min}).

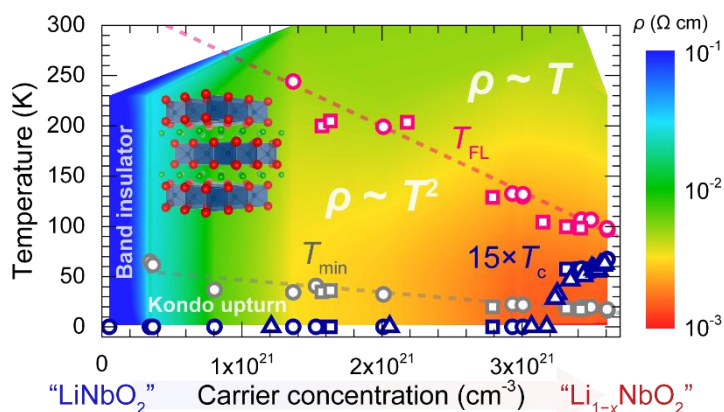


Fig. 1 Electronic phase diagram of Li_{1-x}NbO₂. T_c , T_{min} , and T_{FL} indicate superconducting transition, normal state resistive minimum, and Fermi-liquid transition temperatures, respectively. Circle, Square, and triangle plots correspond to the data obtained from different three electrochemical devices.

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High sensitive magnetic sensors based on anisotropic magnetoresistive effect in epitaxial $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ thin films

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The control of the magnetic anisotropy of ferromagnetic thin films is crucial for device applications, in particular for the proper design of anisotropic magnetoresistive (AMR) sensors. AMR exploits the dependence of the electrical resistance on the orientation of the current with respect to the magnetization in the material. In this work epitaxial $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (LSMO) thin films were deposited on vicinal SrTiO_3 (STO) substrates by pulsed laser deposition in order to induce an uniaxial magnetic anisotropy with an easy magnetic axis along the step edge directions [1]. Gold contacts were deposited by ion beam and Wheatstone Bridges were patterned in order to get rid of common perturbations, such as temperature drift (Figure 1-a).

The effect of the LSMO film thickness, of the angle of the vicinal substrate and of the deposition temperature on the measured magnetoresistance at 310 K were investigated. We will present results obtained with AMR sensors patterned in 30 to 60 nm thick epitaxial $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (LSMO) thin films, deposited on 4° , 6° or 8° vicinal SrTiO_3 (STO) substrates. The detectivity of a magnetic sensor is defined as the ratio of its intrinsic noise to its sensitivity. As shown in figure 1-b), the lowest measured detectivity at 310K was $1.5 \text{ nT}\cdot\text{Hz}^{-1/2}$ at 1 Hz and $200 \text{ pT}\cdot\text{Hz}^{-1/2}$ at 1 kHz in a single-layer sensor without using any performance enhancing techniques such as magnetic flux focusers or modulation techniques [2]. If the signal to be measured is in the low frequency region (i.e. below 100 Hz) as in biomedical applications, such AMR sensors become very competitive when compared to Tunnelling Magnetoresistance (TMR) and Giant Magnetoresistance (GMR) technologies, due to lower $1/f$ noise, small size and much simpler fabrication process [3].

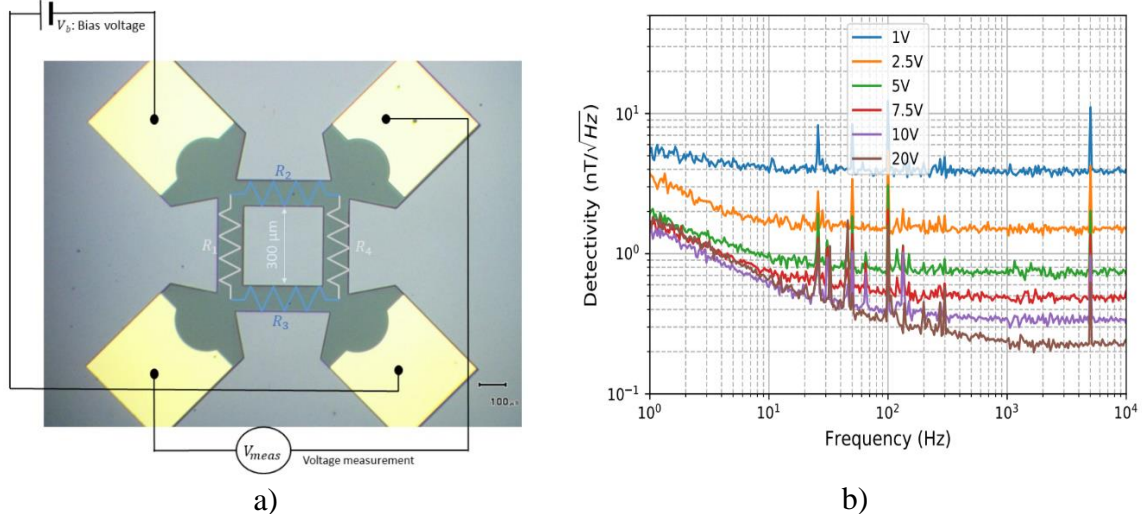


Figure 1: a) Optical microscope photograph of a Wheatstone bridge patterned in LSMO thin films with $300 \mu\text{m}$ long and $100 \mu\text{m}$ wide arms; b) Measured detectivity versus frequency on optimized LSMO AMR sensors at 310 K versus applied voltage

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Poster Session P1

P1-1	Utilizing capping layers to preserve properties of correlated oxide thin films Cohen, Amit (Technion – Israel Institute of Technology Haifa)
P1-2	Phase-separated nanostructures formation of Ni-doped SrTiO ₃ studied using low-temperature scanning tunneling microscopy and spectroscopy Liu, Yen-Po (Forschungszentrum Jülich)
P1-3	Exploring new substrates for the thin film growth of rutiles Birkhölzer, Yorick A. (Cornell University)
P1-4	Fabrication and structural characterization of Nd ₂ NiO ₄ thin films Maritato, Luigi (Università Degli Studi di Salerno and CNR-SPIN)
P1-5	Deposition of spinel IGZO films on optimized GZO templates Agiannis, Evangelos (KU Leuven)
P1-6	Integration of perovskite based transparent conducting oxides on industrial substrates: the key role of the glass substrate properties and binary oxides seed layers Mezhoud, Moussa (Normandie University)
P1-7	Strain-induced phase transition in epitaxial fluorite oxides De Luca, Gabriele (ICMAB-CSIC, Barcelona)
P1-8	Growth by pulsed laser deposition of SrVO ₃ thin films for optical applications Angeloni, Tancredi Thai (University of Geneva)
P1-9	Surpassing the limits: Is it possible to achieve high-purity SrRuO ₃ Thin Films using PLD? Monteiro Cunha, Daniel (University of Twente)
P1-10	Integrating epitaxial barium titanate on various substrates for silicon nitride photonics Shanker, Ojas (University of Twente)
P1-11	Optimizing the growth parameters for stoichiometric CaIrO ₃ thin films Sarantopoulos, Alexandros (Forschungszentrum Jülich)
P1-12	Growth of barium titanate thin films with ozone-assisted ALD Pulikkottil Dinesh, Dharsana (University of Oslo)
P1-13	CO ₂ laser substrate heating at very high temperatures during the deposition of functional films in UHV systems Stein, Wolfgang (SURFACE systems + technology GmbH + Co KG)
P1-14	Monophase in high-entropy oxides as solid-state electrolyte in thin film batteries Das, Shaona (TU Darmstadt)
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[P1-1]

Utilizing capping layers to preserve properties of correlated oxide thin films

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The correlated oxide SrVO₃ attracted significant attention in recent years for its potential in electronic devices and optoelectronics devices. Additionally, it is an end member of the La_{1-x}Sr_xVO₃ system, which exhibits a metal-to-insulator transition, which is interesting for various fundamental and device purposes.

For any practical device, SrVO₃ and related alloys have to be grown in the form of a thin layer, which is prone to surface overoxidation in ambient conditions. To protect the layers, we capped them *in-situ*, and assessed the cap effectiveness in preserving the underlying layer. A combination of spectroscopic methods was employed, combined with cluster calculations; these enabled resolving of the V 2p photoemission spectrum.

We employed both x-ray photoelectron spectroscopy (XPS) and angle-resolved hard x-ray photoelectron spectroscopy (HAXPES) on *in-situ* TiO₂-capped SrVO₃ thin films [1]. The chemical state of vanadium was found to be consistent throughout the sample's depth. This consistency led to the conclusion that the bulk properties of SrVO₃ are preserved up to the top-most few atomic layers adjacent to the cap.

This work lays the foundation for utilizing SrVO₃ and related oxides in practical electronic and optoelectronic devices.

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Phase-separated nanostructures formation of Ni-doped SrTiO₃ studied using low-temperature scanning tunneling microscopy and spectroscopy

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Transition metal doped SrTiO₃ is of significance for devices in energy [1], information [2-3], and catalysis [4]. With the growth characterization of Ni-doped SrTiO₃ (Ni:STO), spontaneous formation of separated nano-phases is observed. In view of controlling the formation of such nano-phases and in a wider context for vertically aligned nanocolumn (VAN) engineering [5], our focus is on the growth mechanism of STO with Ni dopant enrichment in designated areas of the perovskite lattice, associated to the occasional formation of nano-phases embedded in a Ni-doped STO matrix.

Scanning tunneling microscopy and spectroscopy (STM/S) are unique surface techniques for mapping both precise topography and electronic properties down to the atom scale, making them a promising instrument for studying nanostructures.

In this study, we investigate the reflection high-energy electron diffraction-controlled pulsed laser deposition (PLD) grown sub-monolayer Ni:STO on Nb-doped SrTiO₃ (Nb:STO) substrate using low-temperature (LT) STM/S at 4.6 K in ultra-high vacuum (UHV). Upon in situ transfer to the LT-STM, the as-deposited samples with less than 1 u.c. show surface morphology with Ni:STO islands with trenches between the islands. Surprisingly, the specific patterns that can correspond to the nanocolumn patterns on a thick film, similar to VAN morphologies observed in the literature, [6] already appear in less than one monolayer deposition. Island thickness of about 4.1 Å (lattice constant of STO~3.9 Å) is observed on all the sub-monolayer Ni:STO samples, in addition, homogeneously dispersed nanoparticles with 2-3 nm diameters, in consistency with the STEM data, exhibit low-density distribution above the Ni:STO islands.

Local electronic properties of the nanostructures are investigated by STS and dI/dV, yielding the local density of states. A bandgap of about 3.3 eV is seen on the Ni:STO islands (tip-induced band bending takes part in the STS measurement), while the nanoparticles show a slightly larger bandgap, 3.7 eV, attributed to be NiO, furthermore, there is an extra sharp state at 1.2 eV above the Fermi level, indicating increased p-doping cooperated to the STO film.

The study presents the very early stage formation process and the dopant distribution of the embedded nano-phase Ni:STO, allowing for improved control of such nanostructures, likely also relevant to understanding the nucleation of VANs.

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[P1-3]

Exploring new substrates for the thin film growth of rutiles

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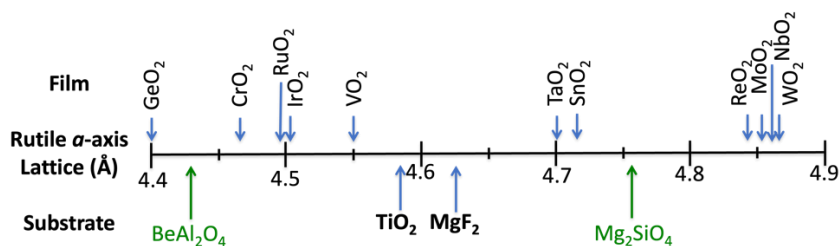
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The synthesis of epitaxial oxide thin film materials with crystal structures other than perovskite is often severely limited by the lack of suitable substrates. A prime example of such materials are binary transition metal oxides with the rutile structure such as CrO₂, VO₂, NbO₂, and TaO₂. In fact, there are currently only two commercially available substrates, TiO₂ and MgF₂, with the rutile structure. The figure below illustrates the lattice parameters of selected rutiles and highlights the need for new substrates with distinct lattice parameters.

Another major challenge for the stabilization of TaO₂ and other exotic binary oxides with rutile-like polymorphs, e.g., WO₂, ReO₂, and MoO₂, is preventing the unwanted oxidation of such desired rutile structures into a more oxidized version of the binary oxide (e.g., WO₃, ReO₃, and MoO₃) due to oxygen transfer from the substrate. To achieve the targeted rutile structure of the thin film, growth must take place at very low oxygen partial pressures, e.g. < 10⁻⁸ Torr in the case of TaO₂. Viable substrates to stabilize such rutile thin films must hence be able to withstand aggressively reducing conditions at temperatures frequently exceeding 1000 °C, dramatically limiting the number of candidate materials. We note that TiO₂ and MgF₂ are very prone to reduction and evaporation, respectively, at such high temperatures, and can react with these desired rutiles. Therefore, despite being the only commercial rutile substrates, they are of limited use for the growth of many rutiles irrespective of their lattice mismatch.

One of the key structural motifs of rutiles is their edge-sharing oxygen octahedral network along the *c*-axis. Here, we explore the use of non-rutile materials that share this structural motif with rutiles. The two candidate materials we selected – Mg₂SiO₄ (known as the mineral forsterite) and BeAl₂O₄ (known as chrysoberyl or alexandrite) – crystallize in the olivine structure. As a first example, we show the epitaxial stabilization of TaO₂ thin films grown by suboxide molecular-beam epitaxy on Mg₂SiO₄. Second, we demonstrate the epitaxial growth of RuO₂ on BeAl₂O₄.



Number line of rutile *a*-axis lattice parameter and two minerals with the olivine structure having similarity in structural motif with rutile.

Fabrication and structural characterization of Nd₂NiO₄ thin films

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In obtaining performing solid oxide fuel cells at intermediate temperatures (IT-SOFC) one of the major limiting factors is the reduced efficiency of the cathode reaction and sloppy kinetic. The use of materials with improved ionic conductivity can mitigate such limitations [1–3]. To this respect, the class of mixed ionic electronic conductors of the type Nd₂Ni_{1-x}Cu_xO_{4+δ} represents a good candidate for innovative cathode materials in IT-SOFC [4]. We have deposited thin films of Nd₂Ni_{1-x}Cu_xO_{4+δ} with $x = 0$ on LSAT and STO crystalline substrates using different physical vapor deposition (PVD) techniques (RF sputtering, molecular beam epitaxy). The deposited samples have been structurally characterized by XRD, EDS and XPS. The desired stoichiometric phase has been obtained after ex situ in air annealing process at high temperature.

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Deposition of spinel IGZO films on optimized GZO templates

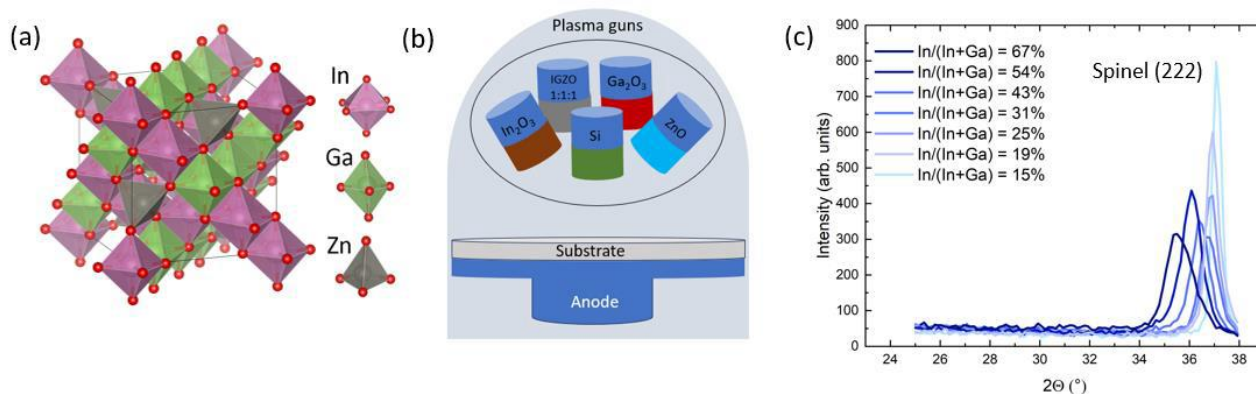
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Indium gallium zinc oxide (IGZO) has attracted much attention as a wide bandgap semiconductor for thin-film transistors (TFTs) due to the absence of mobile holes and the relatively high electron mobility of its amorphous phase [1]. The growth of amorphous films at low deposition temperatures is beneficial for applications as semiconducting channels of TFTs in optical displays and 3D memory elements. Despite its advantages, amorphous IGZO is electrically unstable due to the ease of formation of oxygen vacancies, which act as n-type dopants. Spinel IGZO is characterized by improved oxygen stability due to its more uniform oxygen bonding and can be obtained by PVD at low temperatures [1]. Gallium zinc oxide (GZO) crystallizes only into the spinel phase, with tetrahedral and octahedral sites being occupied by zinc and gallium atoms, respectively. In spinel IGZO (Figure a), the gallium is partially replaced by indium, improving carrier mobility due to the enhanced overlap of its 5s orbitals. Unfortunately, with high indium concentrations the spinel phase becomes structurally unstable.

Using GZO as a template, spinel IGZO can be obtained by PVD at conditions that are normally employed for amorphous films. We explore the deposition of spinel IGZO films with different indium concentrations by co-sputtering from multiple targets that are positioned around the substrate (Figure b). Compositions were measured using the x-ray fluorescence (XRF) technique. As the indium concentration decreases, the x-ray diffraction (XRD) spinel (222) peak shifts to higher values of 2θ , indicating a decrease of the crystal lattice constant. The addition of O_2 to the Ar sputtering gas and the type of power (pulsed DC or RF) are important parameters to enable the creation of spinel IGZO with high indium concentrations.



(a) The crystal structure of spinel IGZO represented by the cubic space group $Fd\bar{3}m$ (227) with interchanging In and Ga atoms. (b) A multi-cathode chamber with 5 targets. The substrate is positioned at the center. Different types of power generators can be used for each target. (c) θ - 2θ XRD of spinel IGZO films with approximately 33% Zn that were deposited using a pulsed-DC with IGZO 1:1:1 target in combination with Ga_2O_3 , In_2O_3 and ZnO targets, using RF PVD.

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[P1-6]

Integration of perovskite based transparent conducting oxides on industrial substrates: the key role of the glass substrate properties and binary oxides seed layers

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The discovery of conductivity and transparency in strongly correlated metals of the ABO_3 type has paved the way for the development of a new generation of indium-free transparent conducting oxides (TCOs). $SrVO_3$ (SVO) perovskite oxide has demonstrated comparable electrical and optical properties compared to Indium-Tin-Oxide (ITO) [1], the standard material for TCO devices. Integrating these new TCOs materials on low-cost substrates for large-scale production remains a technological challenge because of the necessity of a compatible growth template for the crystalline growth of this new generation of TCOs, in contrast to ITO, which can be grown easily on industrial substrates and yet has suitable properties while being amorphous.

In this presentation, we introduce an innovative approach to the integration of SVO thin films onto industrial substrates, such as glass or silicon. Our method uses a TiO_2 seed layer to facilitate growth and enhance crystalline quality of SVO films. The resulting thin films are polycrystalline in nature, exhibiting remarkably low electrical resistivity of about $400 \mu\Omega \cdot cm$ and good optical transparency exceeding 70%. These characteristics underline the immense potential of these films for a wide range of electronic applications. Furthermore, our study also focuses on the intricate interplay between the chemical and thermal properties of the substrate, specifically glass, and their influence on the properties of SVO thin films. A deeper understanding of these substrate properties not only contributes to optimizing the growth process but also highlights the critical role of the substrate in achieving the desired properties of these new TCOs materials.

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Strain-induced phase transition in epitaxial fluorite oxides

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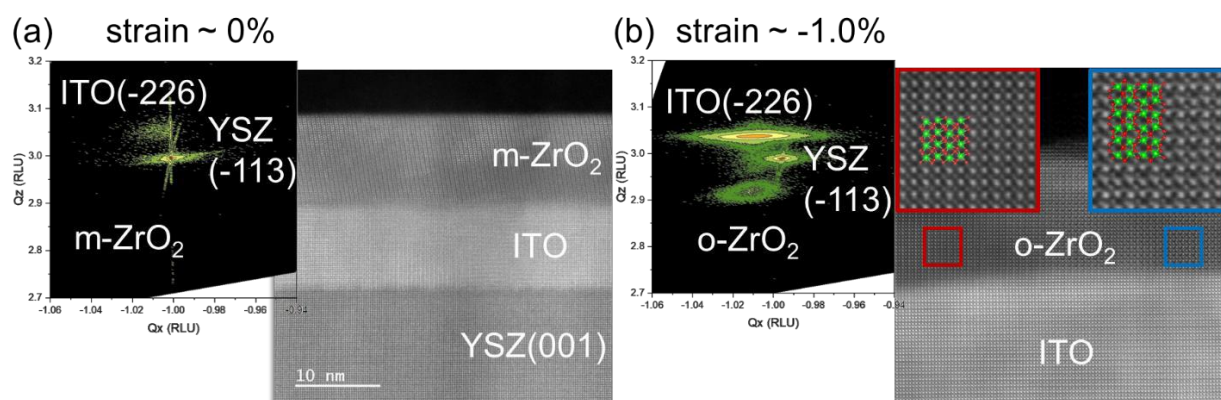
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Epitaxial oxide thin films are crucial for the development of future technologies whose functional properties must be precisely manipulated at the atomic scale. This control enables the improvement of devices' performance, addressing the emerging challenges of our society. While perovskite and related families (Ruddlesden-Popper, brownmillerite, double perovskites, etc.) are ubiquitous in the complex-oxides research area, exploring the nanoscale features of other crystal families is a less-traveled path. One pivotal example is the unforeseen discovery of ferroic properties in fluorite $\text{Hf}_{1-x}\text{Zr}_x\text{O}_2$, an unpredicted result which is now driving the progress in both digital and neuromorphic low-energy-consuming computation [1].

Here, with the aim of developing an epitaxial platform to investigate fluorite (and related) families of oxides, we employed radio-frequency magnetron sputtering and pulsed laser deposition to grow epitaxial thin films of a prototypical fluorite oxide (ZrO_2) on various pristine and buffered single-crystalline substrates, inducing different lattice mismatches. Various parameters (film thickness, growth temperature, buffer layer) affect the resulting ZrO_2 polymorph, with compressive strain inducing a structural phase transition (bulk-like monoclinic to polar orthorhombic) in the strained fluorite film, consistent with early DFT calculations [2]. These results suggest that the methodologies developed in the perovskite oxide community to engineer the nanoscale properties of epitaxial films can be successfully extended to fluorite and related families (pyrochlores, bixbyites) of oxides.



Reciprocal space maps around YSZ (-113) ($a_{\text{YSZ}} = 0.513$ nm) and STEM for $\text{ZrO}_2/\text{ITO}/\text{YSZ}$ heterostructures. Increasing compressive strain induced by the relaxed buffer layer ($a_{\text{ITO}}/2 = 0.506$ nm) drives a transition from bulk-like monoclinic (a) to orthorhombic (b) resulting in ZrO_2 structural domains with the polar axis lying in the film plane.

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[P1-8]

Growth by pulsed laser deposition of SrVO₃ thin films for optical applications

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Light-matter interaction can be strongly enhanced by confining the electric field in optical cavities. These require a well-suited stacking of reflecting and transparent materials selected for the frequency range of interest. In our study, we target the Terahertz spectrum and have chosen the SrVO₃ compound for its high reflectivity in this frequency range [1]. We report results on the growth of SrVO₃ thin films by pulsed laser deposition unraveling the complex dependence of resistivity and crystalline quality on the Ar/O₂ growth atmosphere as well as laser fluence and target-substrate distance [2]. The investigation of electric transport reveals the role of electron-phonon coupling in the conduction of this material, in line with recent literature [3]. Optical measurements performed by Fourier Time-domain InfraRed spectroscopy show that the films reflectivity window is within the scope of our applications.

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Surpassing the limits: Is it possible to achieve high-purity SrRuO₃ thin films using PLD?

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Attaining high residual resistivity ratio (RRR) values in SrRuO₃ (SRO) thin films is a critical goal for enhancing their electronic properties, particularly for applications in oxide electronics. However, ruthenates are very sensitive to defects. The main techniques for the growth of SRO thin films are Molecular Beam Epitaxy (MBE) and Pulsed Laser Deposition (PLD). While MBE has successfully produced SRO films with RRR values close to triple digits (with recent reports showing SRO on DyScO₃(110) with RRR above 200) [1,2], PLD thin films have struggled to surpass single-digit values, with only a single report of RRR of 14.1 for the SRO/NdGaO₃(110) system [3]. Despite the advantage of growing films with consistent quality, the conditions within the plume make SRO films grown by PLD inherently Ru-poor [4]. This research systematically investigates the growth conditions of SRO films on TiO₂-terminated SrTiO₃ (STO) substrates using PLD, focusing on optimizing parameters to achieve higher RRR values.

For this study, SRO thin films were deposited via PLD, with adjustments made to deposition temperature, oxygen partial pressure, and substrate orientation. The films' crystallinity was assessed using X-ray diffraction (XRD), while their electrical properties, particularly resistivity, were measured to determine overall quality and purity. Although high crystallinity alone did not ensure higher film quality, certain conditions showed promise in achieving double-digit RRR values. Notably, films grown on SrTiO₃(111) achieved an RRR of 10.4, as illustrated by the resistivity plot in Figure 1. Through systematic growth studies, this research highlights the potential to overcome PLD's traditional limitations in producing high-quality SRO films. By identifying the critical factors that influence film quality, our findings provide a pathway for further optimization and potential breakthroughs in the fabrication of SRO thin films for advanced electronic applications.

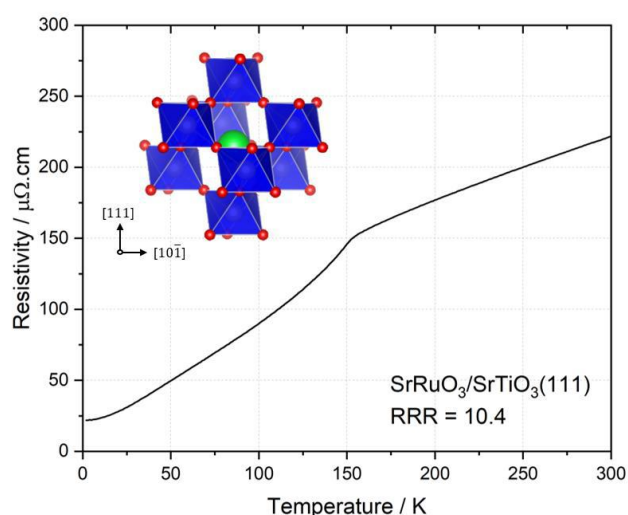


Figure 1. Resistivity vs. Temperature for the SRO/STO(111) system, highlighting its RRR value and its crystal structure.

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Integrating epitaxial barium titanate on various substrates for silicon nitride photonics

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The study of thin film Barium Titanate (BaTiO_3) is critical for advancing the current state of photonic devices for communication technologies. Furthermore, realization of these devices can help provide efficient solutions in the fields of quantum and high-performance computing. BaTiO_3 is a leading candidate in achieving high modulation speeds at low powers due to its exceedingly high linear electro-optic (EO) coefficient, also known as the Pockels coefficient. Due to the anisotropic nature of the EO effect, developing highly crystalline barium titanate with its c-axis in plane (and hence, its polarization vector) is crucial to utilize the highest value of the Pockels coefficient. The initial challenge is to promote growth of in-plane oriented BaTiO_3 while minimizing the formation of out-of-plane oriented BaTiO_3 . Further challenges involve addressing temperature-based integration incompatibilities of BaTiO_3 with current state of the art photonics utilizing silicon nitride waveguides.

Our research explores the epitaxial growth of BaTiO_3 using pulsed laser deposition on three different substrates in parallel as alternative strategies for integration on Si_3N_4 . One strategy is to integrate BaTiO_3 (Tetragonal, $a = b = 0.399$ nm, $c = 0.4035$) [1] on MgO (Cubic, $a = 0.421$ nm) in order to apply in-plane tensile stress and promote in-plane oriented growth, with the additional dual purpose of MgO acting as a lower refractive index material for greater light confinement in the EO BaTiO_3 layer. Another option is to integrate BaTiO_3 on silicon using a 10 nm buffer layer of Strontium Titanate (SrTiO_3). The SrTiO_3 buffer layer induces an in-plane compressive stress which initially promotes out-of-plane oriented BaTiO_3 . However, by tuning oxygen pressure during growth, we have successfully promoted the relaxation and re-orientation of BaTiO_3 from out-of-plane to in-plane. Finally, we attempt deposition of BaTiO_3 on amorphous substrates using $\text{Ca}_2\text{Nb}_3\text{O}_{10}$ nanosheets deposited via the Langmuir-Blodgett method as a template layer to promote epitaxial growth [2]. This is crucial for sidestepping temperature-based monolithic integration incompatibilities, as the BaTiO_3 layer is grown directly on the waveguide or cladding material. Moreover, mode confinement optimization simulations have been performed as a function of various cladding and BaTiO_3 film thicknesses, along with simulations aimed at extracting optimal embedded waveguide geometry.

Ongoing research involves drawing links between tetragonality of the BaTiO_3 thin films and its ferroelectric properties. Subsequently, measurements regarding the electro-optic behaviour and related parameters such as the half wave voltage and propagation loss exhibited by BaTiO_3 thin films on a silicon nitride platform will be performed, enabling steps towards high speed, low power EO modulators.

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Optimizing the growth parameters for stoichiometric CaIrO₃ thin filmsA. Sarantopoulos¹, D. Gustin², O. Concepción³, M. Lezaic^{2,4}, and R. Dittmann¹¹ Peter Grünberg Institute (PGI-7), Forschungszentrum Jülich GmbH, Jülich 52425, Germany² Peter Grünberg Institute (PGI-9), Forschungszentrum Jülich GmbH, Jülich 52425, Germany³ Peter Grünberg Institute (PGI-1), Forschungszentrum Jülich GmbH, Jülich 52425, Germany⁴ Institute for Advanced Simulations (IAS-1), Forschungszentrum Jülich GmbH, Jülich 52425, Germany

5d transition metal oxides, where correlations and spin-orbit coupling play at the same energy scale, create a great playground in search of high-T_C and unconventional superconductivity, spin liquids and novel magnetic ordering phenomena. The metastable perovskite phase of CaIrO₃ was previously grown in thin-film form and characterized as semi-metallic by ab-initio calculations [1], although the optical conductivity spectra were inconclusive in this respect, leaving open the possibility of a small gap insulating state. First-principles calculations yield a small insulating gap, despite a large spatial extent of Ir 5d wavefunctions, and ordering of higher-order magnetic multipoles is shown to go hand-in-hand with the gap formation.

Studying the existence of magnetic multipoles experimentally requires the stabilization of the perovskite phase of high quality CaIrO₃ thin films. The main challenge to this goal arises by the high volatility of IrO₃, due to which, significant off-stoichiometry is observed despite the formation of a stable perovskite structure. Many efforts have been made to stabilize the metastable perovskite structure, which was confirmed experimentally mainly through structural characterization, without adequately addressing the stoichiometric composition of the films. To obtain stoichiometric CaIrO₃ thin films, we had to adjust the growth kinetics while reducing the oxygen partial pressure to prevent the formation of volatile IrO₃. In addition to the stoichiometry control, the deposition parameters were adjusted aiming to improve the crystalline quality and the surface morphology of our thin films. In this work, we will discuss the structural and transport properties of our films based on the deposition conditions, in pursuit of the correct structural properties that enable the presence of multipolar ordering and gap formation, possibly through structural defects.

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[P1-12]

Growth of Barium Titanate Thin Films with Ozone-assisted ALD

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Barium titanate is a widely researched ferroelectric material with a perovskite atomic structure. The possibility of BTO thin films to be incorporated in the silicon-based micro- and nano-fabrication processes make it highly valuable in a range of electronic and photonic applications such as capacitors, thermistors, piezoelectric devices. The growth of high-quality BTO thin films is crucial for enhancing the performance and reliability of these devices. In this study, we are utilizing the advantages of ALD; conformality on 3D substrates [1], low deposition temperatures (giving strong control of the thermal history) [2], and high cation composition control [3] creating an interesting combination of unique properties to be further tweaked.

The ALD process for BTO involves successive deposition of Ba-O and Ti-O unit cycles, utilizing barium bis(2,2,6,6-tetramethyl-3,5-heptanedionate) and titanium triisopropoxide precursors for Ba and Ti, respectively. Ozone acts as the coreactant for both unit cycles, which serves as an alternative to a traditional water vapor-based approach, considering the hygroscopic nature of barium intermediate [4]. The deposition process for both the binary oxide TiO₂ and complex BTO has been studied with its dependence on process parameters such as precursor exposure time, temperature, and ozone dose. Several characterization techniques are utilized to study surface morphology, chemical and structural characterization, and thickness uniformity.

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CO₂ laser substrate heating at very high temperatures during the deposition of functional films in UHV systems

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Epitaxial layer growth of functional films has always a unique and relatively narrow temperature band in which the film grows. Common heating systems such as resistance heating or radiant heating interact with the substrate surface during heating and can cause contamination that has a lasting impact on the layer quality, especially since the heating surfaces are significantly larger than the substrate surfaces [1].

This problem can be avoided if the substrate is heated directly via an energetic beam source like a laser. Laser heaters allow this direct heating, but the substrate must absorb the laser wavelength as completely as possible. SURFACE has been supplying such laser heating systems for over 20 years, usually with diode lasers, emitting in the NIR wavelength range. Direct sample heating requires an absorption of the substrate in the laser wavelength range.



Fig.1: CO₂ laser heater installed at a PLD system

Unfortunately, perovskite substrates, which are often used in the functional film deposition, are highly transparent in this wavelength range, but they absorb in the long wavelength range above approx. 5 μm , which is very well suited by the CO₂ laser with its 10.6 μm . SURFACE has designed and built such systems since 2013 already. Results are reported [2].



Fig.2: 5x5mm substrate at 1600°C

One advantage of CO₂ laser heating is the very large temperature range (>1600°C) and, with a well adapted design, the extremely limited heating of the holding elements. This means that even with a sample size of 5x5mm, the sample holder can be placed on an Omicron stainless steel sample carrier. This structure enables still RHEED measurement during deposition and after the end of the process, the sample holder can be easily transferred in vacuum to enable further layer examinations using a variety of surface analysis tools. This ensures the greatest possible cleanliness in the subsequent examinations.

Summation of the advantages of CO₂ laser heating [2]:

- **Cleaner:** no side contamination of parasitic hot surfaces, no silver paste
- **Faster:** lowest thermal mass, high ramp rates, insitu surface termination at high temperatures
- **Safer:** no HF necessary to terminate the substrate surface

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Monophase in high-entropy oxides as solid-state electrolyte in thin film batteries

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The demand for energy storage devices is on the rise, driven by increasing needs in both stationary and mobile applications. This has intertwined batteries deeply with modern lifestyles. Within the battery community, Li-ion solid-state batteries stand out as the most popular choice due to their ability to discharge at high energy densities (around 10 mA cm^{-2}) and maintain high-power outputs, even under elevated temperatures like 100°C . Despite these advantages, Li-ion batteries face significant challenges related to their electrolytes. These include short lifespan, sensitivity to temperature fluctuations, and cost considerations. Particularly in microelectronics, conventional solid electrolytes often lack the necessary ionic conductivity required for ultrafast charging and discharging cycles. As example LiPON, the most common electrolyte with ionic conductivity $\sim 2 \times 10^{-6} \text{ Scm}^{-1}$ at room temperature, is associated with the highest internal resistance of the cell.

In the pursuit of materials exhibiting high ionic conductivity, recent attention has turned to high-entropy oxides (HEO) featuring a rock salt structure with a minimum of five cations. In 2016, Bérardan *et al.* reported significant findings on the HEO $(\text{Mg, Co, Ni, Cu, Zn})_{1-x}\text{Li}_x\text{O}$, demonstrating an impressive ionic conductivity of up to $1 \times 10^{-3} \text{ Scm}^{-1}$ at 20°C [1]. This discovery opens up possibilities for utilizing this material as a solid-state electrolyte, a development eagerly anticipated [2]. Building on this foundation, efforts have been made to further enhance the ionic conductivity of $(\text{Mg, Co, Ni, Cu, Zn})\text{O}$ through additional doping. This oxide has also been recognized in literature for its notable potential to function effectively as anodes with optimal doping concentrations [3]. Previous studies have mainly employed solid-state synthesis, solution combustion, or nebulized spray pyrolysis methods, all requiring quenching as a crucial step in the material synthesis process. In contrast, limited literature is found on thin films grown by pulsed laser deposition [4]. Here we report a study of the synthesis conditions for monophase $(\text{Mg, Co, Ni, Cu, Zn})\text{O}$ to investigate the change in structure due to different growth kinetics.

We deposited $(\text{Mg, Co, Ni, Cu, Zn})\text{O}$ onto (100)-oriented MgO substrates using different cooling rates of 10° , 40° , and $50^\circ/\text{min}$, while maintaining substrate temperatures around 400°C and fixed p_{O_2} at approximately 50 mTorr. X-ray diffraction (XRD) analysis was used to check for additional peaks corresponding to constituent element oxides near the main substrate peaks. For films grown with faster cooling rates, XRD indicates that they are frozen in the desired entropy stabilized state at room temperature, reducing the likelihood of evolving into multiple phases within the host matrix.

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Decoupling of substrate and epitaxially grown Y123 thin films by $\text{Ca}_2\text{Nb}_3\text{O}_{10}$ nanosheet templates

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The growth of high-quality superconducting ReBCO thin films, notably $\text{YBa}_2\text{Cu}_3\text{O}_7$ (Y123), is highly dependent on the substrate. To enable epitaxial growth, choosing a substrate that matches the lattice parameter of the orthorhombically structured Y123 is critical. The manufacturing of commercial ReBCO tape is performed on an MgO buffer layer [1], while for thin film growth of ideal model systems i.e. perovskite single crystals such as SrTiO_3 , are used as substrates [2]. The necessity to use these types of growth templates for both fundamental research and applications provides the opportunity for alternatives, where superconducting ReBCO can be grown in high quality on any arbitrary substrate. For this a flexible method is needed, where epitaxy in the film is completely independent of the type of substrate used. Realising this will provide new opportunities for the utilization of ReBCO materials like Y123.

Our research demonstrates, the successful epitaxial growth of Y123 thin films on amorphous substrates using pulsed laser deposition (PLD). Contrary to the growth on single-crystalline substrates, here, the growth template is provided by a layer of flexible, crystalline $\text{Ca}_2\text{Nb}_3\text{O}_{10}$ nanosheets [3]. These nanosheets are deposited on amorphous substrates by Langmuir – Blodgett method. Nanosheets are one-unit cell thick 2D structures with sheet sizes around $200\mu\text{m}^2$. On the nanosheets, domains of epitaxial Y123 are realized. All the domains display out-of-plane direction uniformity but in-plane orientation diversity. Moreover, grain boundaries are formed between the domains acting as resistive barriers, which decrease the film's superconducting performance [4]. Yet, by controlling the geometry of the nanosheets, the domain sizes are dramatically enlarged, and the in-plane mosaic twist is reduced. This results in the realization of Y123 thin films with a critical temperature (T_c) of zero resistance of 85K (figure 1) which is close to the standard of 90K for Y123 thin films [2].

Ongoing research entails a detailed investigation of the electronic properties of the Y123 thin films. More precisely, the characterization of the critical current (I_c) and its dependence on the nanosheets due to the still-existing grain boundaries and in-plane differences between domains reduce the critical current [4]. To fully characterize the Y123 films, we are using an ensemble of X-ray, electrical transport techniques, and imaging techniques, which include AFM and SEM. The deposition of nanosheets by the Langmuir–Blodgett method is scalable, flexible and applicable on a wide range of substrates. This provides opportunities of direct superconductor growth on large area (wafer-scale) and curved surfaces. Achieving this opens possibilities for small superconducting devices, like patterned coils.

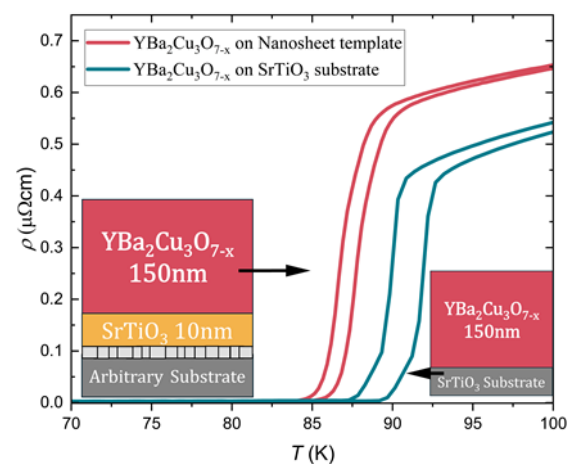


Figure 1: Resistivity versus temperature of superconducting Y123 thin films on different growth templates; nanosheets (red) and single crystal SrTiO_3 substrate (blue).

This research is in collaboration with VDL ETG T&D B.V., The Netherlands.

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[P1-16]

Engineering chiral magnetism in a centrosymmetric oxide

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Perovskite strontium ferrite, SrFeO₃, hosts a variety of chiral magnetic phases at low temperature including multi-q states of different proper screw and/or cycloid ordering [1]. Among them is a phase believed to support topologically-protected magnetic structures and may explain an observed finite-field anomaly in Hall effect [2].

SrFeO₃ is a fascinating material because, unusually, macroscopic chirality exists despite the centrosymmetry of the crystal structure. Instead of a Dzyaloshinskii-Moriya interaction, the helimagnetism has been suggested to arise due to an interplay of electronic interactions [3]. The prospect of helimagnetism that is not borne by the underlying chemical lattice is very appealing from a spintronic applications perspective as it may allow macroscopic chirality switching without movement of atoms.

Using resonant soft x-ray scattering and neutron diffraction, we study these complex magnetic orderings at low temperature on epitaxial thin films of SrFeO₃ grown by pulsed laser deposition. We find that the thin film environment qualitatively alters the magnetic order and we suggest that the character of the chiral magnetism depends on the biaxial strain.

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Band renormalization in LaTiO₃ thin films approaching the Mott transition

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A long-standing problem in the field of correlated electrons is the evolution of the microscopic electronic structure, i.e., the momentum-resolved spectral function, across the Mott transition as the band filling changes [1]. The Brinkman-Rice scenario [2] of a strongly renormalized Fermi liquid predicts a divergence of the effective mass of the quasiparticle excitation, leading to a narrowing of the band width and a decreasing Fermi velocity, when approaching the Mott transition from the metallic side. LaTiO₃ is a prototypical Mott insulator with a 3d¹ configuration, for which we have previously shown that thin films can be *p*-doped by incorporating excess oxygen via adjusting the O₂ partial pressure during pulsed laser deposition [3]. In this way, the films can be tuned from the Mott insulating to the correlated metal phase. In addition, irradiation with X-rays, as in photoemission, can desorb apical oxygen at the film surface, allowing finetuning of the band filling in terms of *n*-doping [4]. In all cases, the exact band filling can be determined from tight-binding fits to the measured band dispersions. Here, we study the Mott transition by investigating the renormalization of the quasiparticle band along a high-symmetry direction upon band filling, using angle-resolved soft X-ray photoemission spectroscopy. We observe an increase of the effective mass as the Mott transition at half-band filling is approached, with a stronger increase in surface-sensitive measurements at lower photon energy than in more bulk-sensitive measurements at higher photon energy, suggesting stronger electron correlations at the surface. A quantitative account to the band renormalization is provided by slave-spin meanfield calculations for various values of the onsite Coulomb repulsion, confirming that the surface is more strongly correlated than the bulk.

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Exploring topochemical oxygen insertion in Fe oxides for reversible tuning of thermal conductivity

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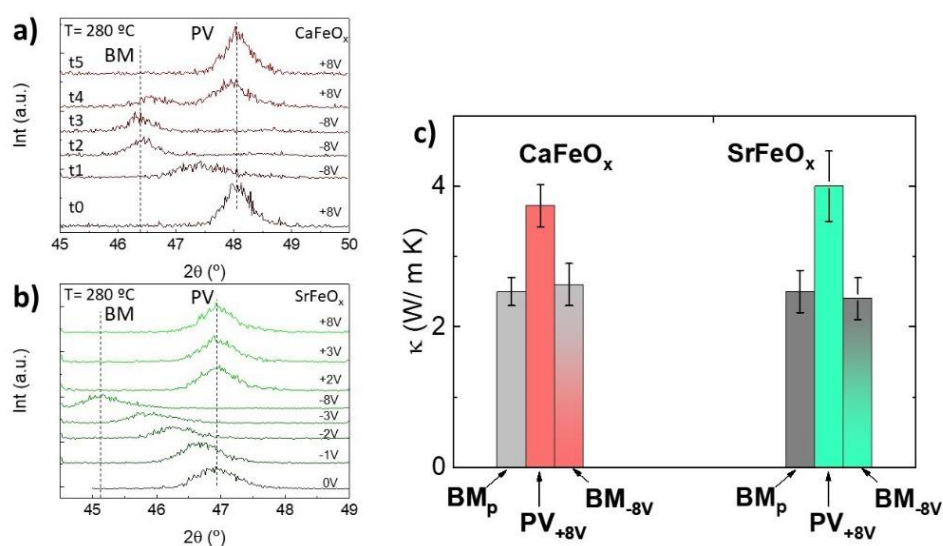
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The development of functional materials whose thermal conductivity can be switched between different states plays a key role in several technologies, such as nanoelectronics or thermoelectrics. Oxygen-deficient ABO_{3-x} perovskites are promising candidates in this direction, since they exhibit fast O^{2-} mobility at high temperature, being possible to induce reversible transitions between perovskite (PV, ABO_3) and brownmillerite (BM, $ABO_{2.5}$) structures at moderate temperatures under oxidizing/reducing conditions.

Here, we report different routes for the BM-PV transformation in $(Ca,Sr)FeO_{3-x}$ thin films, and the effect of the O^{2-} insertion mechanism on the thermal conductivity of the phases and reversibility of the process. In particular, we have explored the use of a gas oxidant (O_2 and O_3), NaClO aqueous solutions, and dry electrochemical oxidation using a solid electrolyte (YSZ). High-resolution transmission electron microscopy and chemical microanalysis demonstrate that liquid phase oxidation of the BM leads to partial amorphization of the structure due to Ca^{2+}/Sr^{2+} leaching, limiting the reversibility of the process. Repeated oxidation in O_2/O_3 atmosphere at high temperature also accumulates lattice defects that produces a progressive loss of cyclability. On the other hand, dry electrochemical oxidation results in full reversibility and high crystallinity of both BM and PV phases, so that the system can be cycled several times without apparent degradation.

Our results demonstrate that liquid-phase oxidation should be avoided in the topochemical oxygen insertion of ferrites, and present solid-state electrochemical oxygen exchange as the best route for reversible tuning of thermal conductivity.



a,b) X-ray diffraction of $CaFeO_{3-x}$ and $SrFeO_{3-x}$ thin films, respectively, during solid-electrolyte electrochemical oxygen insertion. c) Reversibility of the thermal conductivity after BM-PV-BM cycling.

Electrical control of magnon propagation in iron oxide garnet thin films

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The electromechanical and electromagnetic coupling in rare-earth iron garnets (RIG) are very small due to their centrosymmetric structure [1]. To overcome this weakness, we demonstrated RIG thin films of broken spatial inversion symmetry by applying tensile strain that allows the spontaneous coexistence of spin and electric dipole originating from tetragonal distortion. In this work, we fabricated $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG, 12.376Å-bulk) thin films on lattice matched GGG (12.38Å) and 1% lattice mismatched SGGG (12.50Å) substrates by pulsed laser deposition (PLD) technique. Room temperature Fe $L_{2,3}$ X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) measurements (fig.1(a)-(b)) revealed a low orbital moment in YIG/GGG, suggesting minimal crystal distortion, while the high orbital momentum of 0.15 in YIG/SGGG films indicates the quenching of orbital moment sum is alleviated by the reduction of crystal symmetry from cubic to tetragonal. Also, X-ray diffraction (XRD) and reciprocal space mapping (RSM) revealed strained epitaxy indicating that the in-plane lattice of YIG is matched with GGG and elongated to align with SGGG (Fig.1(a)). Here-after, coplanar waveguides (CPWs) and gate electrodes were fabricated by photolithography and sputtering as described in the schematic of Fig.1(d). Spin wave transmission characteristics were measured using vector network analyzer (VNA). Fig.1(e, f) summarizes a systematic shift in spin wave transmission resonance frequency (Δf) versus electric field (E). In tensile-strained YIG (120nm)/SGGG, the transmission spectra showed a notable rightward shift with electric field. In contrast, lattice-matched YIG (120nm)/GGG samples subjected to a high electric field of up to 800 kV/cm exhibit only a minimal response. The SW frequency shift likely arises due to the electrical response of polarized domains within tensile-strained RIG thin films that ultimately alters the magnetic anisotropy of RIG thin films through the strong spin-orbit coupling (Fig1(b)). This simple technique of electrical control of magnons represents a revolutionary advancement in magnonics, potentially paving the way of developing ultra-low power logic devices using spin waves.

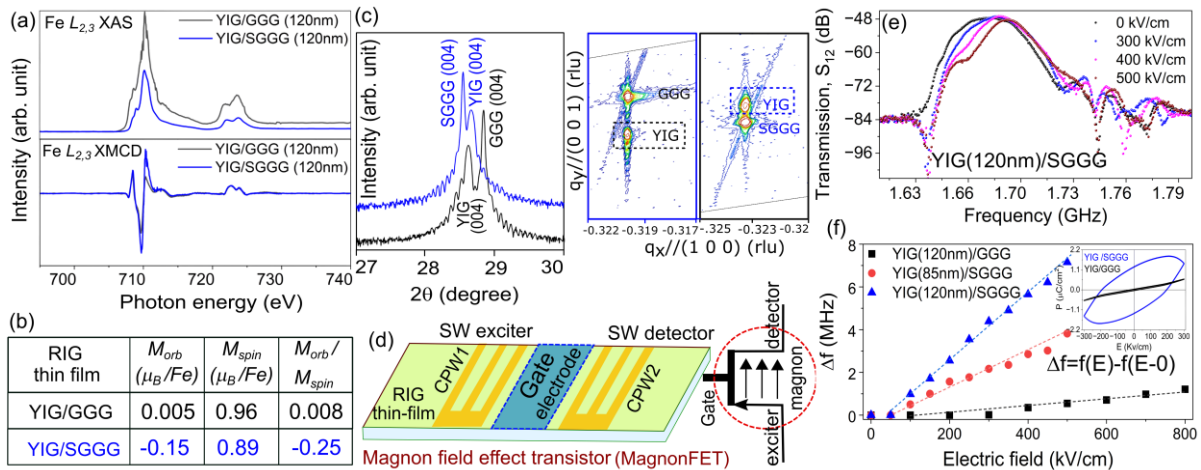


Fig. 1. (a)-(c) Magnetic and structural properties of lattice matched YIG/GGG (001) and tensile strained YIG/SGGG (001) thin films, (a) Fe $L_{2,3}$ XAS and XMCD, (b) summary of magnetic moments, (c) XRD 2θ - ω scan and RSM, (d) schematic of fabricated spin wave devices on RIG films, (e) SW transmission spectral shift with applied E field, (f) SW transmission resonance freq. shift [$\Delta f(E) = f_{res}(E) - f_{res}(E=0)$] versus applied E field (inset: polarization characteristics).

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Impact of interfacial proton accumulation on protonation in SrCoO_{2.5} oxides

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Electrochemical protonation provides facile ways to control physical properties and even explore unprecedented phases of solid-state materials. While how proton accumulation changes materials' properties has been investigated, how protonation and proton evolution in solids can be controlled and promoted remains an enigmatic puzzle. In the work reported here, we investigated the influence of electrochemical proton injection duration (t_{vg}) on the protonation of SrCoO_{2.5} (SCO) films in electric-field-effect transistor structures with gate layers of the proton-conducting electrolyte Nafion. We found that while the total time of proton injection is fixed, the proton concentration accumulated in SCO films varies depending on the duration of the proton injection. When protons are injected in a relatively short t_{vg} (≤ 600 sec), the concentration of hydrogen accumulated in SCO film increases with increasing t_{vg} , reaching the maximum proton concentration of ~ 1.9 per formula unit of SCO for the $t_{vg} = 600$ s case. On the other hand, when t_{vg} is longer than 900 s, the proton concentration no longer increases with t_{vg} but decreases, implying the occurrence of counterreactions that oxidize the channel and extract protons from protonated SCO. These observations indicate that protons accumulated at the Nafion/SCO interface play a significant role in the proton evolution and protonation of SCO films and that controlling the interfacial proton accumulation by adjusting the proton injection duration is the critical factor to enhance proton evolution and maximize the proton concentration accumulated in SCO films.

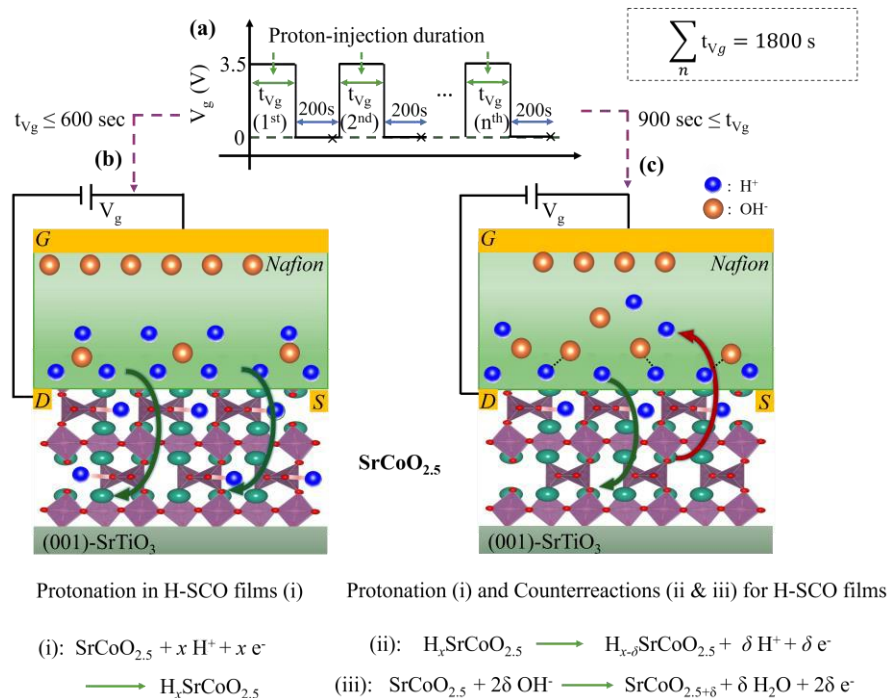


Figure 1. Voltage sequence for protonation of SCO films (a). Schematics for proton evolution and reactions for the cases of (b) $t_{vg} \leq 600$ sec and (c) $t_{vg} \leq 900$ sec

Neutron methods for investigation of hydrogen induced modifications of correlated oxides

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Hydrogen induced modifications of correlated oxide layers have gathered increasing interest in the last decade. On one hand we have several examples of proton-based devices working on the principle of reversible electrical or magnetic switch through hydrogen intercalation and removal, already demonstrated for various oxide systems [1-3]. Another branch of great interest is that of superconducting nickelate systems, in which the role of hydrogen is still under lively debate [4-5]. For all these systems the full understanding of the mechanisms leading to the electronic modification of the oxide is hindered by the difficult detection of hydrogen in low-mass thin films. Neutron reflectometry (NR), in combination with other complementary techniques, can be a powerful tool for non-destructive, in situ, non-local characterization of such systems. Due to the negative scattering length of hydrogen atoms and the possibility to use isotope contrast variation neutrons are a highly sensitive probe to quantify hydrogen in a complex host matrix. In this poster we will illustrate the capabilities of NR to correctly distinguish between distinct processes happening in thin film oxide while reacting with hydrogen-containing substances or gases, such as (i) hydrogen incorporation, (ii) oxygen depletion, and (iii) lattice expansion. We will show its application in several example systems including hydrogen gas exposed manganite and nickelate single layers, as well as CaH₂ and CaD₂ reduced infinite layer nickelate heterostructures (Figure 1).

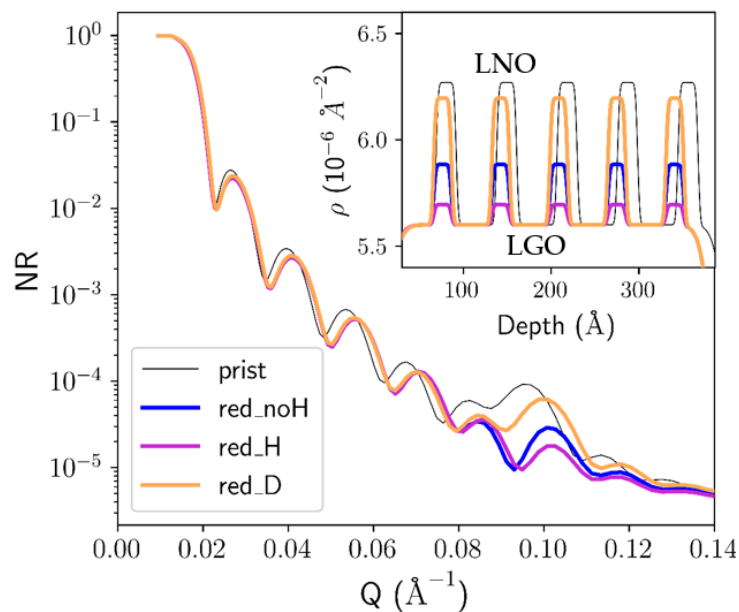


Figure 1 NR simulation of the different signal for a pristine and reduced infinite layer nickelate(LNO)/gallate (LGO) heterostructure with and without hydrogen incorporation

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[P1-22]

Probing the Electronic Structure of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ thin films grown by pulsed laser deposition

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The emergent properties that may arise at interfaces in heterostructures of dissimilar materials are of great interest in the pursuit of novel quantum materials for spintronics applications. One such example is the combination of a topological insulator and a magnetically ordered material, in which the magnetic order serves to break the time reversal symmetry of the topological insulator, creating a gap in the topological surface state. As components in such a system we consider thin films of the topological insulator Bi_2Te_3 and the ferromagnetic oxide $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$.

As a step towards understanding the electronic structure of such a heterostructure, we must first understand the electronic structure of its constituents. Here, we consider epitaxial thin films of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ grown by pulsed laser deposition on (111)-oriented SrTiO_3 substrates. We first develop an in-vacuum methodology for surface preparation of the films by annealing in an oxygen atmosphere, enabling us to measure the electronic structure of films that have been exposed to air. The methodology is accompanied by X-ray photoelectron spectroscopy to study the surface stoichiometry, and low energy electron diffraction to study the surface crystallinity. The electronic band structure of the films is probed by soft X-ray angle-resolved photoemission spectroscopy using a synchrotron light source.

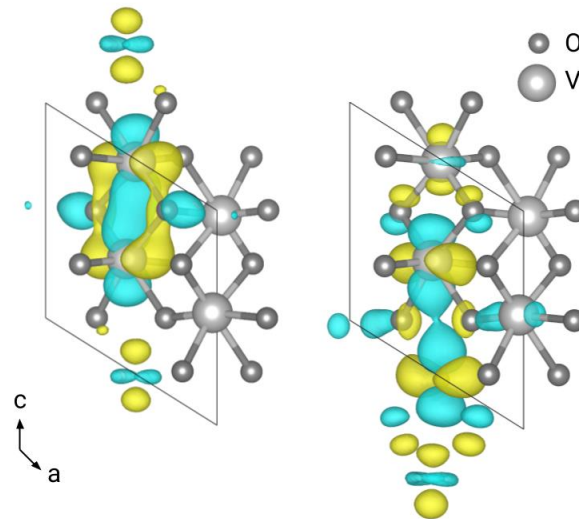
Finally, the experimental results are compared to band structures obtained from density functional theory calculations.

Single-site DFT+DMFT for vanadium dioxide using bond-centered orbitals

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We present a combined density-functional theory and single-site dynamical mean-field theory (DMFT) study of vanadium dioxide (VO_2) using an unconventional set of bond-centered orbitals as the basis of the correlated subspace [1]. VO_2 is a prototypical material undergoing a metal-insulator transition (MIT), hosting both intriguing physical phenomena and the potential for industrial applications. With our choice of correlated subspace basis, we investigate the interplay of structural dimerization and electronic correlations in VO_2 in a computationally cheaper way compared to other state-of-the-art methods such as cluster DMFT. Our approach allows us to treat the rutile and M1 monoclinic VO_2 phases on an equal footing and to vary the dimerizing distortion continuously, exploring the energetics of the transition between the two phases. The choice of basis presented in this work hence offers a complementary view on the long-standing discussion of the MIT in VO_2 and suggests possible future extensions to other similar materials hosting molecular-orbital-like states. Our approach is also pivotal for future calculations of VO_2 with larger system sizes to accommodate dopants or for studying strain dependence of the system.



Some of the bond-centered orbitals used in this work for the M1 phase of VO_2 . V (O) atoms are shown in light (dark) gray and yellow (blue) isosurfaces at identical values for all plots show the positive (negative) phase of the orbital.

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Light-matter interactions in spin-orbit-entangled transition metal systems

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Jaume Gázquez¹, Gervasi Herranz¹

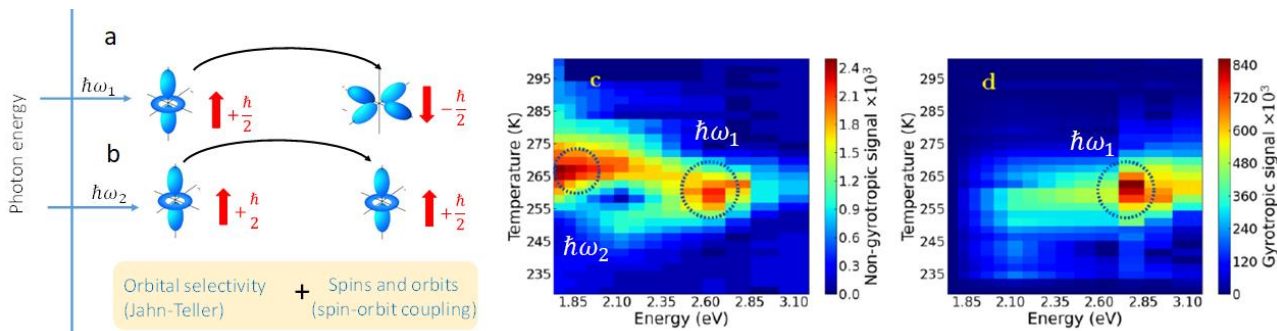
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The excitation of different degrees of freedom (DoF) and their mutual interactions offer an excellent platform to investigate fundamental physics in strongly correlated systems and pave the way to enhanced functionalities. One interesting perspective is to excite a particular DoF (spin, orbital or lattice) and probe how it couples dynamically to other DoFs. Here we consider the coupling of states with specific orbital symmetry to lattice modes and spin excitations in Mn^{3+} d -orbital states in manganite oxides³. In the latter, Jahn-Teller interactions promote orbital polarization, which allows to drive resonant photoexcitations between specific orbital symmetries (see Figures a – c) [1,2]. Interestingly, we show that high-spin $t_{2g}^{\uparrow\uparrow\uparrow} e_g^{\uparrow}$ and low-spin $t_{2g}^{\uparrow\uparrow\downarrow} e_g^0$ configurations become spin-orbit entangled, whose entanglement is detected by a resonance in magneto-optical maps (Figure d). This allows to probe the dynamics of orbital/lattice and spin/orbital coupling through ultrafast spectroscopies. Our results allow a new perspective on the use of optical spectroscopies to analyze the dynamical coupling of different DoF in a large family of materials, including manganites and their extremely rich phase diagram (charge/orbital order, complex magnetic phases, metal-insulator transitions). In such approach, optical spectroscopies may be used as a complementary tool to other spectroscopic techniques used in correlated systems which are orbital-selective, e.g., resonant inelastic x-ray scattering (RIXS).



Two resonance photoexcitations involving end states of different orbital symmetry are depicted in a) and b). Panel c) maps the optical absorption as a function of temperature and photon energy, where resonances $\hbar\omega_1$ and $\hbar\omega_2$ are detected. In contrast, magneto-optical maps in d) only detect resonance $\hbar\omega_2$.

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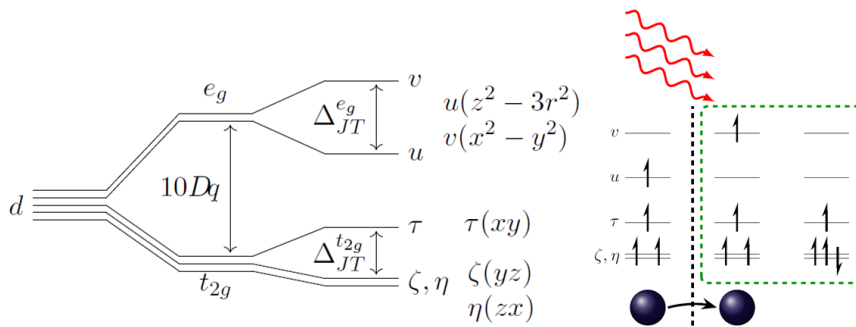
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Non-equilibrium approach to light-matter interactions in spin-orbit-entangled d -electron systems

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Large spin-orbit coupling and strong electron correlations in d -electron systems are an excellent platform to study fundamental physics in condensed matter. This class of materials host a number of correlated phases, including spin-orbit Mott insulators, (quantum) spin liquids, multipolar orbital order, hidden phases, etc. Coupling to electromagnetic fields offers an interesting approach to study excitations in these systems. Interestingly, such photoexcitations involve the interplay of different degrees of freedom (DoF), e.g., spin, lattice or orbital DoFs. Here we focus on the development of theoretical tools based on non-equilibrium many-body physics to study the interaction of electromagnetic fields with t_{2g} - e_g states in transition metal compounds. One of our interests is in analyzing possible orbital contributions to the transient and steady-state responses to electromagnetic fields. In particular, we focus on Jahn-Teller interactions in the e_g -manifold, which allows to analyze resonant photoexcitations between states of specific orbital symmetries, and their coupling to lattice modes and spin excitations. We use our approach to study photoexcitations in Mn^{3+} d -orbital states (as found in manganite oxides), a prototypical Jahn-Teller ion, where high-spin $t_{2g}^{\uparrow\uparrow\uparrow}e_g^{\uparrow}$ and low-spin $t_{2g}^{\uparrow\uparrow\downarrow}e_g^0$ configurations may become spin-orbit entangled, whose entanglement is detected by resonances in magneto-optical maps [1, 2, 3]. From this formalism we obtain transient and steady-state current densities and optical conductivities for a range of values in parameter space (exchange energy, crystal field, spin-orbit coupling, vibronic interactions). We also include a discussion about the extension of the model to other transition metals with different d^n configurations and values of parameter space. Our work highlights the relevance of using electromagnetic fields to probe and manipulate correlated phases in d -electron systems.



We investigate t_{2g} - e_g states interacting with electromagnetic fields, taking into account contributions from spin-orbit coupling and Jahn-Teller interactions. The latter promote spin-orbit entanglement of high- and low-spin configurations (dashed box on the right), which is detected by magneto-optical spectroscopy [1-3].

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- [3] A. S. Miñarro et al., *submitted*.

Possible orbital momentum accumulation in the light early transition metal oxide SrVO₃

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The emergence of transporting orbital currents in materials with low spin-orbit coupling (SOC), along with the ubiquitous existence of orbital texture in different material classes, represent groundbreaking electronic phenomena in solids, holding promise for fast and energy-efficient electronics.[1, 2] Pioneering experiments by D. Lee et al. [3] evidenced the possibility to exploit orbital torque in different normal metal (NM)/ferromagnetic (FM) heterostructures, despite the possible presence of spurious effects hindered full understanding the orbital momentum accumulation. Recent advancements, such as those by Choi et al. [4], definitively established orbital currents in a simple device using a single-element thin titanium film, revealing a twentyfold amplification in orbital momentum compared to its spin counterpart. Ferromagnetic resonance (FMR) experiments NM/ FM systems serve as a robust tool for exploring orbital transport in solids. Rooted in theoretical work by Tserkovnyak et al. [5], a direct proportionality between the damping constant (α) of the FM layer and the NM density of states (DOS) has been established. As corroborated through experimental validation in spin-based systems,[6] this relationship provides a powerful strategy for modulating interfacial spin transport properties within magnetic devices. Extending the latter considerations to orbital transport at the NM/FM interface, we propose using SrVO₃ (SVO) thin films, a low-SOC early transition metal oxide, as an orbital current generator. Manipulating SVO's DOS and orbital occupancy through strain conditions on selected substrates is feasible.[7] Here, we reported FMR- based studies on different SVO(15 nm)/FM(i.e. Ni, Co) heterostructures, where the SVO is under different epitaxial-strain conditions. Notably, an enhancement of α is observed when the SVO layer is introduced, with respect to a bare FMR reference, further exhibiting a dependence on the lattice mismatch compared to the substrate. Unexpectedly, the less strained SVO/Ni system showed a α value comparable with the one extracted from the high-SOC Pt/Ni bilayer, supporting the possible existence of orbital accumulation in the SVO thin film. The acquisition of second harmonic magnetoresistance on the same SVO/FM systems further supports the substrate-dependent magnitude of measured orbital torque. These results mark an initial exploration of early transition metal oxides for spin-orbitronics, presenting a strategy for optimizing orbital-charge conversion efficiency in real electronic devices.

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Laser-induced quenching of metastability in a V_2O_3 thin film at the Mott-transition

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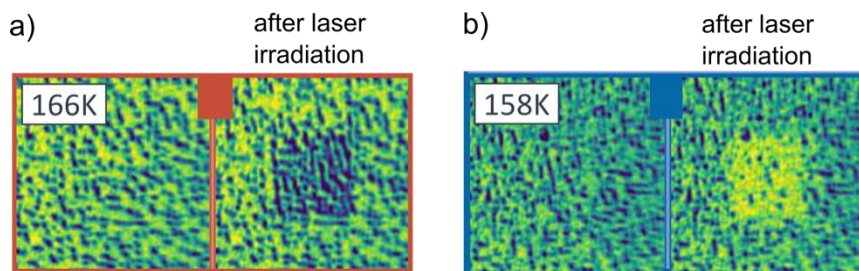
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Strongly correlated insulator thin films are actively investigated in the emerging field of neuromorphic computing. In previous studies on thin films of the prototypical Mott-insulator V_2O_3 , a strain-induced spontaneous separation into insulating and metallic domains during the phase transition was discovered [1, 2]. These domains arrange themselves in a characteristic herringbone pattern. Here, we used low-temperature optical microscopy to investigate the effect of focused laser scanning irradiation [3]. We found that the response depends on the thermal history. Because the phase transition is first-order, the V_2O_3 thin film has a thermal hysteresis. Laser scanning predominately induces metallic domains if the thin film is heated to a temperature in the insulator-to-metal transition interval on the heating branch. This behavior can be expected because the laser heats the thin film locally, which might drive some domains from the insulating to the metallic state. However, laser scanning predominately induces insulating domains if the thin film is cooled to a temperature in the metal-to-insulator transition interval on the cooling branch, which cannot be explained by simple laser heating. We conjecture that the observed effect is due to the thermal metastability of the first-order Mott transition, and that the laser spot acts as a local perturbation that drives the superheated or supercooled thin film into a stable state.

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Photomicrographs of herringbone domains in a V_2O_3 -thin film. a) acquired at the insulator-to-metal transition on the heating branch of a thermal cycle. Laser scanning irradiation induces metallic domains. b) acquired at the metal-to-insulator transition on the cooling branch. Laser scanning irradiation induces insulating domains.

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Kinetics and reversibility of ion-triggered metal-insulator transitions in complex oxides

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Complex oxides have evolved as a major class of functional materials applied in a wide range of energy conversion and storage approaches as well as electronic devices, which harvest the ability to precisely tailor and combine oxides on the nanoscale. Here, miniaturization and confinement phenomena determine the ionic-electronic structure of oxide thin films, surfaces, and interfaces.

Moreover, complex oxides can often undergo intrinsic transitions in electrical conductivity which can vary by orders of magnitude or even show true metal-insulator transitions depending on their specific defect structure. A prime example for this is reflected by the perovskite-to-brownmillerite phase transition in cobaltite oxides, which is accompanied by an ion-triggered metal-to-insulator transition.

Here, we will discuss how iontronics can be exploited to trigger structural, electronic, and magnetic phase transitions [1] and discuss means to control the dynamics of oxygen exchange in nanoscaled oxides. [2] As we demonstrate for the model thin film oxide (La,Sr)CoO₃, the structural transition from perovskite to Brownmillerite takes place in an already insulating and non-magnetic reduced perovskite phase. In contrast, the order-to-disorder transition in the oxygen-deficient phase is not responsible for the observed metal-to-insulator transition. Based on ambient pressure XPS and in-situ structural characterization, we reveal the oxygen-surface exchange to reflect the rate limiting step in the reaction kinetics. Enrichment of A-site cations close to the surface are responsible for a proceeding degradation of the thin films under repeated cycling in oxidizing and reducing environments.

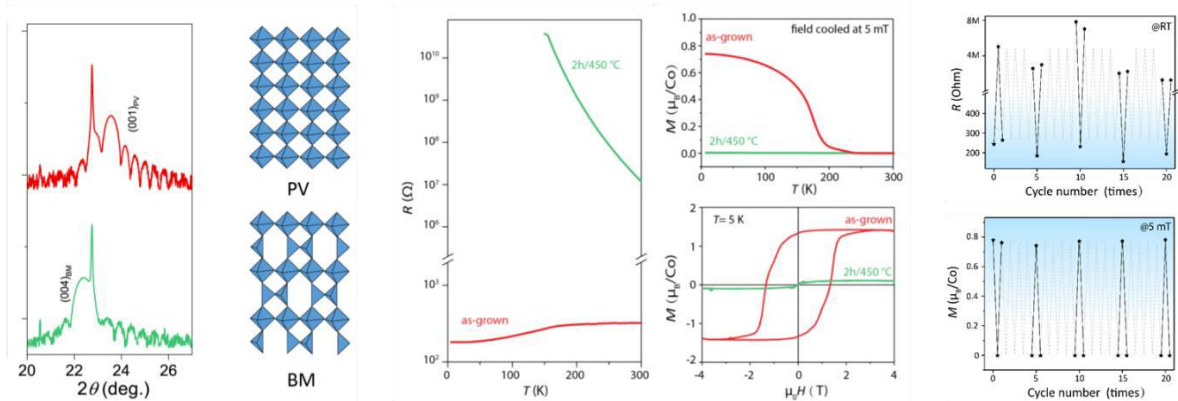


Fig. 1: Reversibility of the Perovskite-to-Brownmillerite phase transitions.

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Large spin and orbital antimagnetolectricity in BiCoO₃

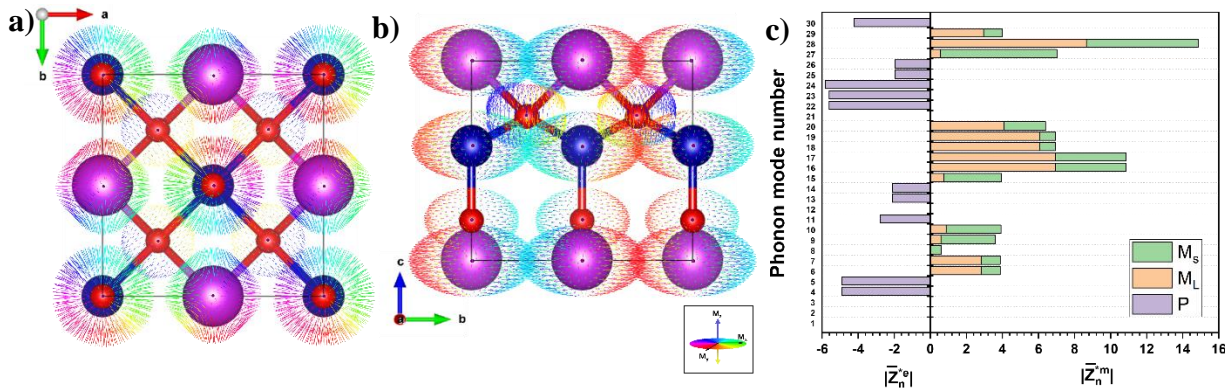
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BiCoO₃ (BCO) crystallizes in the P4mm space group at 870°C and 6 GPa, exhibiting a super-tetragonal phase ($c/a = 1.267$) that allows for a giant polarization of approximately 180 $\mu\text{C}/\text{cm}^2$. The Néel temperature of BCO is reported to be 470 K [1]. Recent experimental findings [2] have indicated a non-zero linear ME response in C-AFM phase - the most stable - yet prohibited by symmetry. This unexpected discovery motivated our investigation into the underlying mechanisms driving this phenomenon.

Using first-principles calculations and symmetry analysis, we explore the magnetoelectric properties of the room-temperature multiferroic crystal BCO. We use applied magnetic field, finite-difference techniques and density functional perturbation theory (DFPT) to show that BCO is actually anti-magnetolectric at the linear level. The calculation of the dynamical magnetic effective charges (DMC) [3] reveals that the total magnetoelectric response is zero due to the compensating non-zero magnetoelectric response of each magnetic sublattice (akin to the hidden order explained in antiferromagnets by Louis Néel). This calculation also highlights that the orbital contribution to the response is remarkably larger than the spin one (See figure c) and giving a rather large total magnetoelectric response of 85 ps/m for each magnetic sublattice. Furthermore, we provide an intuitive recipe to visualize the dynamical magnetic effective charge, which in turn allows examining its multipolar nature which we confirm by means of ab initio calculations (see Figure a, b). Given this large value of the local response, we investigate the ferromagnetic phase as well, which give a giant linear magnetoelectric response of about 1000 ps/m and coming mainly from the spin contribution this time. Finally, we discuss the possible reasons for such a large magnetoelectric response in BCO and propose possible strategies to unveil this potentially large response.



Schematic representation of the DMC pseudotensor as viewed from a) [001] and b) [100] directions. The color of the vectors surrounding Bi (purple), Co (blue) and O (red) atom represent the direction of the induced magnetization when the atom is displaced in a given direction that cross a sphere of arbitrary radius. c) Amplitude of mode Born effective charge (e) (purple) and DMCs (μB) from spin (green) and orbital (orange) of each phonon mode in the C-AFM phase.

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STEM-based in-situ study of electrically induced phase transitions and related phenomena at the nanoscale

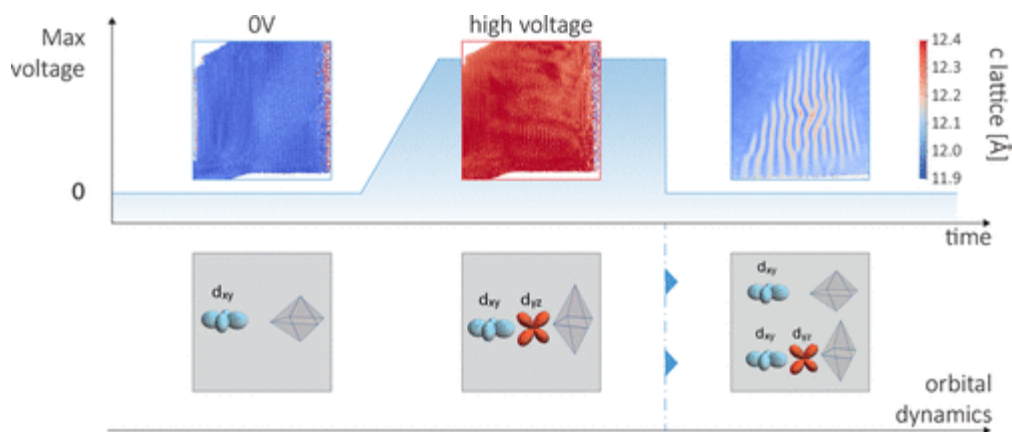
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Over the last years, in-situ biasing experiments inside the transmission electron microscopy (TEM) have been become more readily feasible and have gained great interest. I will discuss a few topics where this methodology was used to follow electric field-induced oxygen vacancy migration and electrostriction in CGO [1,2] and metal-insulator transition (MIT) phase transition under a static electric field in Ca_2RuO_4 [3].

I will discuss the advantages offered by the TEM to apply STEM-based methods such as 4DSTEM, EELS, EDX and ptychography to follow the evolution of structure, chemistry and electronic structure upon such phase transitions and during field application.



Structural phase transition and pattern formation by electric-field quenching in Ca_2RuO_4 single crystal (reproduced from Ref. 3)

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Orbital assisted switching of electronic phases in V_2O_3 thin films

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The exploration of electronic correlations and orbital occupancy across the metal-insulator transition in a strongly correlated V_2O_3 system provides valuable insights into fundamental physics. It also enables the control and enhancement of material functionality, which holds immense importance for diverse technological applications [1, 2]. In the current study, we have explored the essential role of trigonal distortion together with the orbital distributions in iso-structural paramagnetic metal (PM) and paramagnetic insulating (PI) phases of V_2O_3 thin films. We stabilized the isostructural PI and PM states at room temperature (RT) in epitaxial V_2O_3 thin films using RHEED assisted Pulsed laser deposition technique by controlling the degree of trigonal distortion. X-ray linear dichroism (XLD) at the O K-edge is employed to uncover electronic reconstructions in our studied films both in PM and PI states as shown in Figure 1.

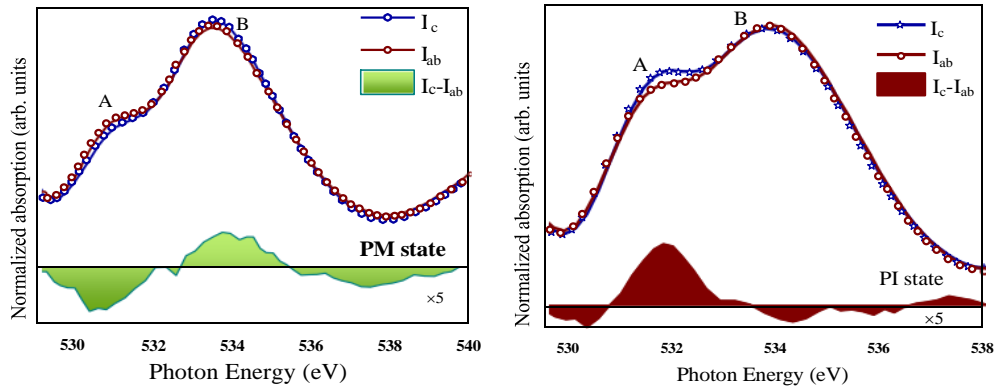
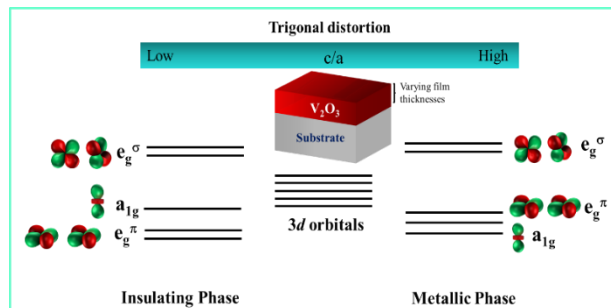


Figure 1. XLD signal at the O K-edge of V_2O_3/Al_2O_3 thin film in PM and PI phases.

Importantly, the PM state demonstrates electron occupation preference in orbitals perpendicular to the system, while the PI state reveals a switch in electron occupation preference within the plane of V_2O_3 thin film (as shown in the Schematic below), indicating an intriguing orbital switching from PM to PI state. Our results demonstrate the orbital switching across PM to PI transition together with an enhancement in electronic correlation strength (U/W) leading to an emergence of a band gap in PI state at RT. These findings deepen the understanding for tailoring the materials properties and promises for advancing material design together with improve the functionality of the system in electronic switching applications [3].



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Bulk electronic structure of SrNbO₃/SrTiO₃ thin films

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The realization of quantum materials (QMs) with both electron correlations and topological properties is currently under active investigation. In this context, 4d transition metal oxides offer a good balance between electron-electron correlations and spin-orbit coupling. Recently, a topological band structure was predicted for an orthorhombic phase of SrNbO₃ with $a^-a^-c^+$ octahedral rotations [1]. In fact, epitaxial strain can control octahedral rotations in SrNbO₃/SrTiO₃(001) (SNO/STO) films breaking the cubic symmetry and generating a novel Dirac semimetallic phase [2]. A thickness-dependent high linear magnetoresistance and high mobilities were reported in this system [3]. The understanding of these unique properties relies mostly on theoretical descriptions of the SNO electronic structure. Until now, only a few experimental studies exist to explore and understand the electronic properties. Although the synthesis of pristine SNO thin films is challenging due to the metastable nature of the Nb⁴⁺ cation with a d¹ configuration, it can be preserved by capping layers. Here, we have grown STO-capped epitaxial SNO/STO thin films of various thicknesses from strained to relaxed conditions using pulsed laser deposition. As capping layers hinder the lab-based XPS investigation of thin film and interface properties because of its high surface sensitivity, we employed bulk-sensitive hard X-ray photoelectron spectroscopy (HAXPES) at beamline P22 of DESY. Nb core level spectra reveal mixed valence states of Nb⁵⁺, Nb⁴⁺ and show an increase in the Nb⁴⁺/Nb⁵⁺ ratio with increasing SNO thickness which is also reflected in the near-Fermi region. Depth profiling at a particular thickness does not show significant changes in the Nb core-level spectral shape demonstrating the homogeneity of the Nb valency in the film. We also discuss the possible origin of multiple peaks in the Nb core-level spectra.

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Lattice distortions and 3d – 4f exchange interactions in RVO₃ thin films

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Oxides, in particular transition metal oxides of the ABO₃ perovskite structure, are of great interest due to their electronic degrees of freedom and interactions. These lead to a wide range of physical properties (insulating, metallic, superconducting, piezoelectric, ferromagnetic...) that can be tailored by tuning chemical pressure and/or lattice constraints, for example. This great versatility opens up the possibility of functionalizing these materials in nanometric heterostructures for new electronic devices. Within this class of materials, we are particularly interested in the rare-earth vanadates RVO₃ (R = La-Lu or Y), whose antiferromagnetic spin ordering temperature (T_{SO}) lies in the (100K-150K) temperature range. At higher temperatures, V 3d t_{2g} orbital ordering is accompanied by a structural transition with decreasing symmetry, from orthorhombic *Pbnm* to monoclinic *P2₁/m* [1]. In addition to VO₆ octahedra rotations, it has been predicted by *ab initio* calculations that rare earth shifts and Jahn-Teller effect could be exploited to introduce improper hybrid ferroelectricity [2].

Previously, it was proposed that, additionally to typical fields, the chemical potential that controls the concentration of ion vacancies can be a parameter to further tune their properties and achieve new functionalities [3,4]. Growing PrVO₃ epitaxial thin films – by pulsed laser deposition – on a SrTiO₃ substrate, we revealed a specific pathway to achieve this effect, which is similar to an uniaxial strain applied along the growth direction [5]. While the 3d electrons are sensitive to the strength of the crystal field, the 4f electrons undergo a large spin-orbit coupling that enhances the magnetic anisotropy, especially in strained films. Exploring the exchange interactions between the moments of the localized R³⁺ 4f electrons and the spin of the more delocalized V³⁺ 3d electrons is thus important for the development of new materials with tailored, novel or coupled electronic and magnetic properties. To address the magnetic and structural couplings in the PrVO₃ thin films, we have selectively probed the chemical and magnetic properties of the V³⁺ and Pr³⁺ sublattices by using X-ray Absorption Spectroscopy (XAS) and X-ray Magnetic Circular Dichroism (XMCD).

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Phonon mediated superconductivity in complex oxidesJulien Varignon¹A. A. Carrasco-Alvarez^{1,2}, L. Iglesias², S. Petit¹, M. Bibes², W. Prellier¹¹ *Laboratoire CRISMAT, CNRS UMR 6508, ENSICAEN, Normandie Université, 6 boulevard Maréchal Juin, F-14050 Caen Cedex 4, France*² *Unité Mixte de Physique, CNRS, Thales, Université Paris Saclay, 91767 Palaiseau, France*

Several oxides deriving from a Ruddlesden-Popper (RP) phase with the chemical formula $A_{n+1}B_nO_{3n+1}$ such as bismuthates $BaBiO_3$ [1] and antimonates $BaSbO_3$ [2] ($n=\infty$), infinite layered nickelates $R_{1-x}A_xNiO_2$ [3] ($n=\infty$, $R=La, Pr$ or Nd , $A=Ca$ or Sr) and $Nd_6Ni_5O_{12}$ [4] ($n=5$), obtained after a chemical reduction of a parent RP phase, or ruthenates Sr_2RuO_4 [5] ($n=1$) are superconductors either in bulk or once appropriately doped with holes or electrons. However, the attractive interaction responsible of the Cooper pair formation at the core of superconductivity in these complex oxides is not yet fully understood and unified between compounds, thereby hindering the identification of high critical temperature T_c superconductors whose record at ambient conditions is still held by cuprates. Using *first-principles* simulations performed with parameter free density functional theory (DFT), we reveal the existence of an insulating phase in the phase diagram of these complex oxides that is associated with a charge/orbital and bond ordered state. The electron-phonon coupling is then weakened by doping effects (or by other stimuli) so that the materials become metallic. Nevertheless, we find that it remains sufficiently large to mediate the Cooper pairs formation with computed T_c 's in sharp agreement with experiments. These results suggest that the phonons responsible of insulating phases in complex oxides [6] may also be at the core of their superconducting state, irrespective of the presence of correlation effects inherent to transition metal elements. Finally, these results are only ratified if all relevant degrees of freedom, such as structural lowering events and local spin formation, as well as an exchange-correlation functional sufficiently amending self-interaction errors inherent to practiced DFT are involved in the simulations.

Acknowledgements

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Manipulating the metal-insulator transition in ultrathin oxide films by strain engineering

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Metal-insulator transitions (MITs) of strongly correlated perovskite oxides have been intensively studied in the past [1-3]. In thin oxide films, numerous phenomena can emerge when the film thickness is reduced towards the 2D limit, e.g., in SrVO₃ (SVO) [4]. In our previous studies on SrTiO₃ (STO) capped films, a transition from the Mott insulating state at 6 u.c. to metallic behaviour at 10 u.c. thickness has been observed [5,6]. As the thickness decreases, spectral weight is transferred from the quasiparticle (QP) peak to the lower Hubbard band (LHB), with the QP weight being completely suppressed and a charge gap opening when the film enters the Mott insulating regime. The hindrance of electron hopping in the ultrathin films reduces the effective bandwidth and consequently results in the domination of short-range Coulomb repulsion. Another way to finely tune the ratio of kinetic energy and local Coulomb repulsion is by applying lateral tensile and compressive strain.

Here, we report on coherently strained SVO thin films grown on various substrates with different lattice constants by pulsed laser deposition (PLD) in an Ar gas background. Using X-ray photoelectron spectroscopy and transport measurements, we find that the MIT in SVO thin films can be finetuned by both film thickness and substrate strain.

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La doping of SrVO₃: tuning the effective mass in a TCO

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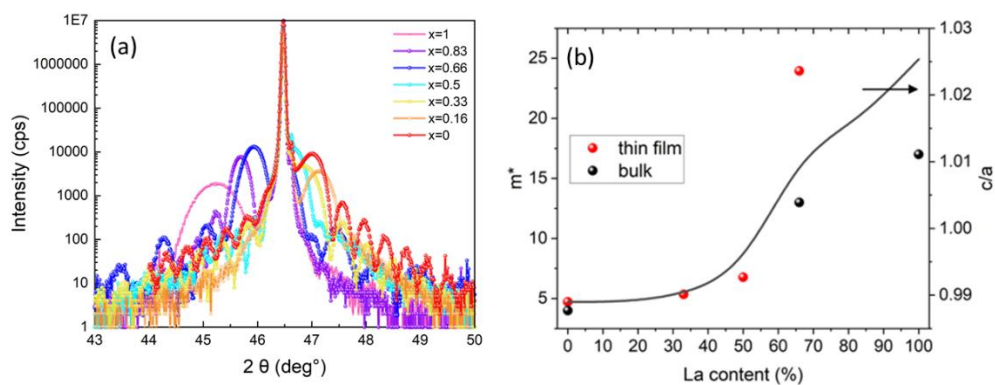
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As a new family of high performance and indium-free transparent conductors (TCOs), the perovskite vanadates have gained a strong interest in the recent years [1]. Due to its V⁴⁺ oxidation state, the prototypical SrVO₃ is a high charge density n-type metallic conductor, with moderate charge mobility and strong electronic correlations. We have studied the La-doping of this material in thin film form in order to explore its effect on the TCO properties, but also the interplay between the enhanced charge density and the lattice distortion on the electronic correlations. A special focus was put on the identification of the charge carriers, as the other parent compound of this system, LaVO₃, is known as a p-type conductor.

We have prepared monocrystalline thin films on SrTiO₃ (001) substrates by Pulsed Laser Deposition, covering the full composition range by forming the solid solution in-situ through the deposition from the two parent compound targets. The films show high crystalline quality, and the typical Metal-Insulator-Transition around $x = 0.8$. No p-type conduction was found in the metallic region of the solid solution, indicating that this state is limited to defect conduction in the insulating phase. The effective mass shows a strong relation to the strain state of the film, allowing to stabilize a low effective mass state for films doped up to 50% La, and an enhanced mass for films above. Therefore, the key parameter for both of the main functional properties of a TCO, optical transparency and the electrical conduction, can be effectively tuned by the strain state.



(a) X-Ray Diffraction patterns of the La_xSr_{1-x}VO₃ thin film series, and (b) a comparison of the evolution of the effective mass (circles) and the film lattice distortion (line) throughout the solid solution

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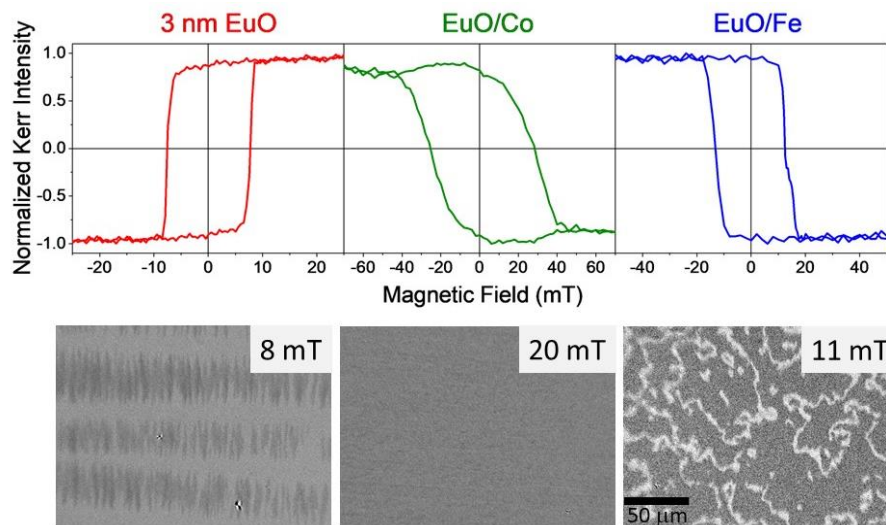
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Synthetic oxide ferrimagnets emerging in proximity-coupled EuO heterostructures – a domain structure analysis

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Heisenberg ferromagnet europium oxide (EuO) thin films are perfectly suited as a spin-functional material for spintronic applications [1]. EuO exhibits robust ferromagnetism (FM) with saturation magnetization of $7 \mu_B$ with a Curie temperature (T_c) of 69 K. The latter limits the practical applications and thus pathways to enhance the T_c have been explored such as altering stoichiometry, doping, lattice strain, etc. [2]. Alternatively, it is promising to explore EuO in proximity to a room temperature FM such as Fe or Co to realize an enhancement of T_c in ultrathin EuO [3].

In this study, magneto-optic Kerr microscopy has been employed to investigate the temperature-dependent magnetization in ultrathin EuO films, both isolated and in proximity to 3d ferromagnets (Co and Fe). Simultaneous hysteresis recording and magnetic domain imaging revealed variations induced by the proximity effect [4]. A comparative analysis of the coupling effects of Fe and Co on the magnetic domains and coercivity behavior of EuO was conducted at temperatures below and above T_c . In a recent study, the thickness of the EuO layer is found to be crucial, indicating that the observed antiferromagnetic proximity coupling is a short-range and genuine interface phenomenon. This fact turns the bilayer into a strong synthetic ferrimagnet [3]. Understanding the microscopic changes induced by the proximity effect could significantly contribute to developing future spin-functional devices utilizing EuO films.



Hysteresis in ultrathin EuO film, EuO/Co, and EuO/Fe heterostructure measured at 30 K. The bottom row shows corresponding domain images observed at different values of the applied magnetic field near the coercive field.

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2D magnetic order in EuO-based synthetic ferrimagnets - induced by proximity coupling

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Magnetic proximity effects make it possible to adjust ferromagnetic properties in an experimentally simple and physically elegant way: If two ferromagnetic films are brought into contact, the electronic coupling changes their magnetic exchange interaction across the interface. For a strong 4f magnet with low T_C , such as EuO, which is coupled to weaker 3d FM with high T_C , such as Fe or Co, even magnetism at room temperature is possible [2]. Moreover, magnetic proximity coupling does not change the stoichiometry or the insulating properties of EuO.

We present MCDAD-HAXPES experiments (Beamline P22, PETRAIII) on the element-specific magnetism of an EuO film proximity-coupled to a 3d FM overlayer using magnetic circular dichroism (MCD) in angular distribution (AD) [1]. The angle-dependent signal provides the in-plane projected magnetization components of Eu 3d and Fe(Co) 2p, and therefore allows a depth profiling of the element-specific magnetic moments. Together with atomistic simulations, we find that the EuO/Fe(Co) proximity coupling is of antiferromagnetic (AFM) nature, and a short-ranged and genuine interface phenomenon. An increase of magnetic order in EuO and of the total T_C of the proximity-coupled bilayer is observed, which is particularly strong in the 2D limit at the interface. The macroscopic magnetic configuration of the EuO/Co(Fe) bilayer in the 2D limit is that of a synthetic ferrimagnet—hence combining advantages of FM and AFM, i.e. easy control of net magnetization and fast spin dynamics.

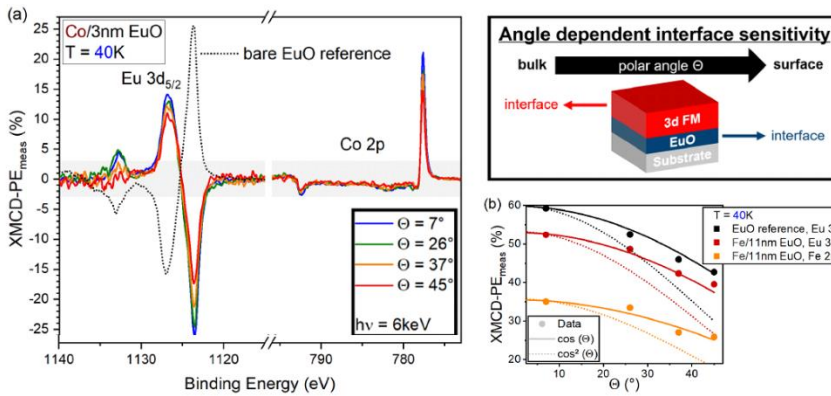


Fig. 1: Angle-dependent MCDAD-HAXPES of the Eu 3d_{5/2} and Co 2p core levels of a 3nm EuO/ 4nm Co sample recorded at $h\nu=6\text{keV}$ and $T=40\text{K}$ at the beamline P22 (PETRAIII). As expected for an in-plane magnetized sample, the MCDAD-HAXPES signal decreases with increasing angle with $\cos(\Theta)$, while at the interface, the Eu-related signal strongly increases with Θ .

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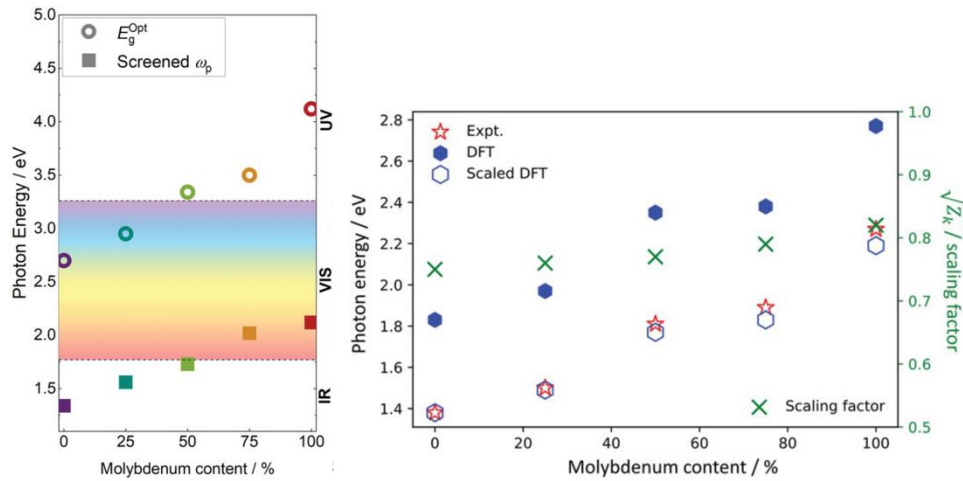
Integrating theoretical and experimental approaches to tailor optical properties in transparent highly conducting perovskites

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SrVO₃ and SrMoO₃ are remarkable conducting d¹ (V) and d² (Mo) perovskites metals with an intrinsically high transparency. In this work, taking the solid-state solution SrV_{1-x}Mo_xO₃ as an example and integrating the theoretical and experimental approaches, we demonstrate how the optical properties of these materials can be engineered for applications as transparent electrodes and in plasmonics, by elucidating how 3d/4d cationic substitution tailors the optical transition energy and plasma frequency. The theoretical electronic structure calculations demonstrates that the shift of the plasma frequency is governed by the interplay of d-band filling and electronic correlations. As a consequence, the absorption and reflection edges of SrV_{1-x}Mo_xO₃ can be shifted to the edges of the visible light spectrum, resulting in a material that potentially outperforms indium tin oxide (ITO) due to its extremely low sheet resistance. Experimentally the optimum has been achieved for $x = 0.5$, where a transmittance above 84% is found in the whole visible spectrum combined with a low resistivity of 32 $\mu\Omega$ cm. In summary, this study advances the knowledge about the peculiar class of highly conducting perovskites towards sustainable transparent conductors and emergent plasmonics.



Left: overview of indirect transition energies and screened plasma frequency. Right: Calculated screened plasma frequency by DFT, and scaled with renormalization factor obtained from DFT+DMFT, in comparison with experimental measurements.

Vanadate TCO on glass substrate using CNO nanosheets as a template: effect of thickness on the film properties

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CaVO₃ and SrVO₃, which are oxide perovskites with strong electronic correlations, are promising and novel transparent conductors [1]. Indeed, the electronic correlations induce an increase of the charge carrier effective mass in those materials, shifting the plasma frequency out the visible range, which induce visible transparency. Therefore, the material's structure and crystallinity are key factor in order to tune their electronic and optical properties.

The ability to integrate vanadates on low cost substrates, particularly glass, is mainly responsible for the technological potential of these new transparent conductors. The growth of vanadates directly on glass is a challenge, as it is not conducting in its amorphous form [3]. Thus, it is necessary to find a growth strategy that preserves the crystalline character of the material. Using a seed layer of [Ca₂Nb₃O₁₀]⁻ nanosheets deposited onto glass substrate, Boileau et al. recently showed that it is possible to integrate crystalline vanadate thin films with competitive properties by pulsed laser deposition [4].

In this work, we study the effect of thickness on the properties of those films. The nanosheets seed layer allow to grow the films at a low thermal budget (500°C) required for the integration on glass. We show the evolution of electrical and optical properties using transport and spectroscopic analyses. Additionally, we probe the local electrical properties of the films by means of scanning spreading resistance microscopy (SSRM).

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Quest for p-type transparent conducting oxides

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Development of p-type transparent conducting oxides (TCOs) has been a challenging issue because the introduction of holes into deep valence bands based on oxygen states essentially tends to break chemical bonds. The known p-type TCOs have been established on subtle balance between advanced crystal growth techniques including doping methods and sophisticated band engineering. Here, we review some of the ongoing approaches that we and other groups have recently implemented with using the pulsed-laser deposition technique.

The first example is a layered oxide $\text{Li}_{1-x}\text{NbO}_2$, where the hole doping associated with Li^+ -ion deintercalation induces not only p-type metallic conductivity, but also superconductivity below 4.2 K [1]. The averaged transmittance to the visible light of 100-nm-thick films is as high as 77%, despite high concentration of hole carriers. Such high transparency can be explained in terms of novel band engineering: largely isolated Nb bands and strong electron correlations.

The second example is one of the widest-bandgap oxide semiconductors Ga_2O_3 . Naturally n-type conductivity of Ga_2O_3 can be readily enhanced by doping donor impurities such as Si. In contrast, p-type conductivity has never been achieved due to deep acceptor levels. Departing from unconventional approaches of making heavily nitrogen-doped films, we show our recent experiments results: partial substitution of oxygen with sulfur in crystalline and amorphous films for making valence bands shallower. NO gas as a source of O as well as N and GaS target as a source of S are used for these experiments.

The final one does not rely on doping but strain engineering for modifying band structures of epitaxial perovskite-type titanates [2]. Owing to a variety of commercially available perovskite oxide substrates, a range of biaxial in-plane strains exceeds $\pm 4\%$. Katase et al. recently demonstrated to change originally p-type LaTiO_3 to by applying compressive stain. Inspired by their achievement, one may expect the opposite change from originally n-type one to p-type. We will discuss this possibility with some of our preliminary results.

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Giant tuning of transport properties and orbital switching by epitaxial strain in Sr-doped LaCrO₃ thin films

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Sr-doped LaCrO₃ (La_{1-x}Sr_xCrO₃) has reemerged as an important functional oxide due to its various outstanding properties, from transparent *p*-type conductivity in the visible range, thermoelectricity and antiferromagnetism among others [1]. In this communication, we will show that high quality La_{1-x}Sr_xCrO₃ thin films can be epitaxially grown by solid-source molecular beam epitaxy (MBE) in the full composition range ($0 \leq x \leq 1$) on different perovskite oxide substrates [2]. For La_{0.75}Sr_{0.25}CrO₃ (LSCO) thin films, yielding the maximum thermoelectric power factor (PF $\sim 0.5 \mu\text{W cm}^{-1} \text{K}^{-2}$) [1,2], we will show that epitaxial strain within $\pm 2\%$ range can greatly tune the transport properties, *i.e.* electrical conductivity (σ) over two orders of magnitude and the Seebeck coefficient (S) by a factor of two (Fig. a), which is explained by modified Cr 3d – O 2p orbital hybridization [3]. Also, we will show that epitaxial strain can preferentially switch the orientation of Cr 3d t_{2g} electronic orbitals from out-of-plane (compressive) to in-plane (tensile) (Fig. b).

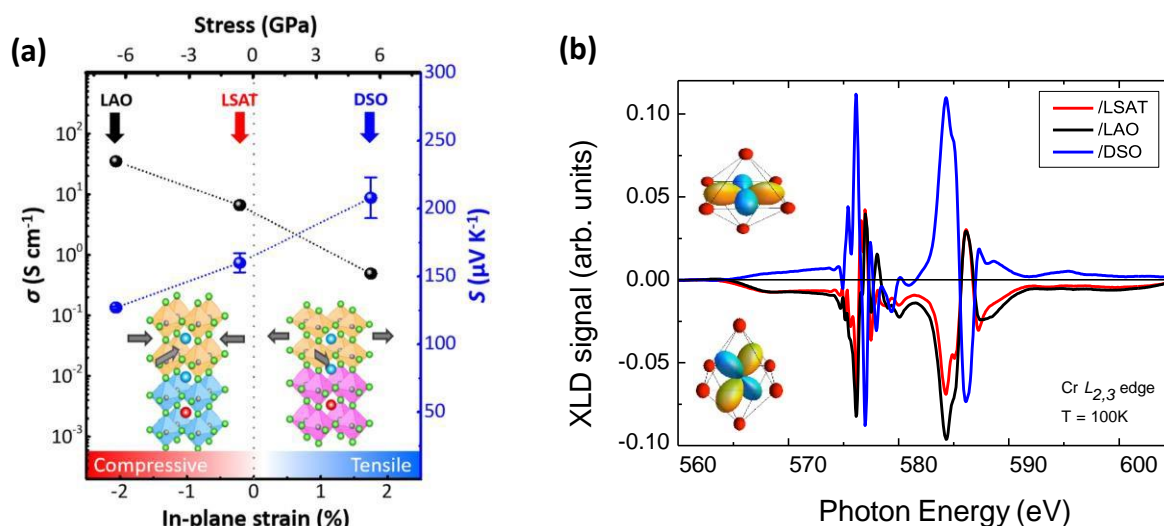


Figure: (a) Electrical conductivity (σ) and Seebeck coefficient (S) measured in-plane on LSCO thin films epitaxially strained on different perovskite oxide substrates (LAO, LSAT and DSO). (b) XLD signal of Cr $L_{2,3}$ edge measured at 60° from normal incidence on the epitaxially strained LSCO thin films.

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Non-reciprocal magnon Hanle effect in antiferromagnetic α -Fe₂O₃ thin films

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Magnons are the quantized excitations of the spin system in magnetically ordered materials and offer a unique platform for future information technology. Antiferromagnetic materials host pairs of spin-up and spin-down magnons, which can be described in terms of a magnonic pseudospin [1]. The precession of this magnon pseudospin about the equilibrium pseudofield, which captures the nature of magnonic eigenexcitations in an antiferromagnet, gives rise to the magnon Hanle effect [2-5]. This effect can be investigated by electrically injected and detected spin transport in antiferromagnetic insulators, demonstrating its high potential for devices and as a convenient probe of magnon eigenmodes and the underlying spin interactions in the antiferromagnet [5].

We investigate the magnon Hanle effect in the electrically insulating antiferromagnetic oxide α -Fe₂O₃ (hematite) via nonlocal magnon transport using two-terminal devices [2-5]. α -Fe₂O₃ harbors a finite Dzyaloshinskii-Moriya interaction together with an easy-plane anisotropy. This results in a slight canting of the sublattice magnetizations in the (0001) plane at room temperature and hence in a residual net magnetic moment [6,7]. Upon cooling, bulk α -Fe₂O₃ exhibits a spin reorientation transition at $T_M = 263$ K from easy plane to easy axis (Morin transition), a feature often absent in thin films. We tune T_M by using different growth conditions, which enables us to investigate the magnon Hanle effect above and below T_M . Unlike for films deposited in molecular oxygen, we observe a finite T_M for those grown in atomic oxygen even down to a thickness of 19 nm [5]. In the easy-plane phase of α -Fe₂O₃ above T_M , we observe a non-reciprocity in the magnon Hanle signal, depending on the magnon propagation direction [4,5]. We explain this observation in terms of an antisymmetric pseudofield along the spin transport direction, which can be controlled by an external magnetic field. The observed nonreciprocal response in hematite films opens interesting opportunities for realizing exotic physics predicted so far only for antiferromagnets with special crystal structures.

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Dirac-like fermions anomalous magneto-transport in a spin-polarized oxide two-dimensional electron system

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The interplay between spin-orbit coupling (SOC), inversion and time-reversal symmetry (TRS) breaking, and hexagonal warping in two-dimensional electron systems (2DES) is the playground for exotic quantum phenomena. By engineering a spin-polarized oxide 2DES with Rashba-like SOC and hexagonal band warping, here we present the first report of an anomalous quantum correction to the magnetoconductance by Dirac-like fermions experiencing competing weak anti-localization and weak localization back-scattering (Fig. 1(a) and ref. [1]), with a phenomenology analogous to that of gapped topological insulators [2]. The results were obtained on the 2DES formed at the epitaxially grown interfaces between (111) LaAlO₃, EuTiO₃, and SrTiO₃ single crystal, characterized by a trigonal crystal field splitting and ferromagnetism induced by Eu and Ti ions magnetic ordering [3]. The anomalous magnetoconductance disappears at the magnetic critical temperature [4], showing a direct link with the ferromagnetic order. The data are explained theoretically in a single band scenario as the combined effects of the Rashba-SOC, of the band-warping induced by the 2DES trigonal symmetry, and of the magnetic gap opening at spin-orbit induced Dirac-like point, giving rise to a non-trivial Berry phase (fig. 1(b, c)). These findings open perspectives for the engineering of novel spin-polarized functional 2DES holding promises in spin-orbitronics and topological electronics.

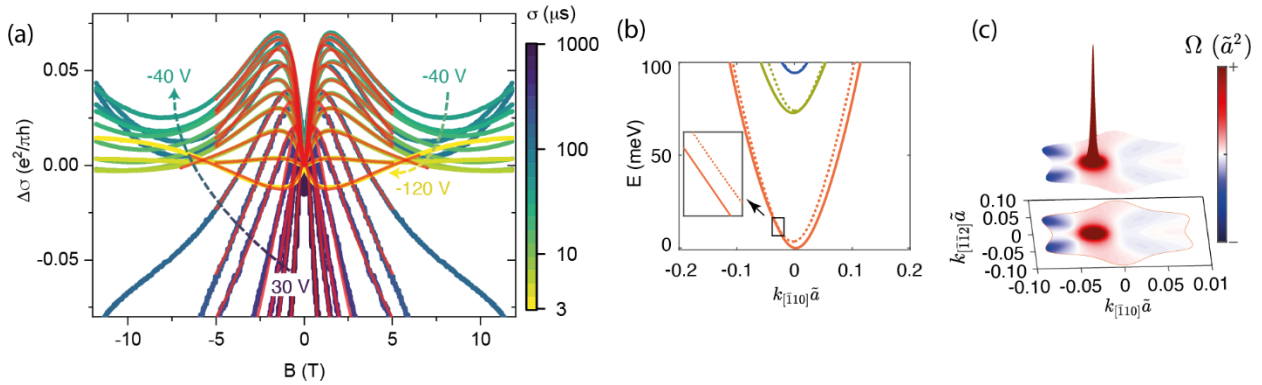


Figure 1. (a) Gate dependence of the anomalous magnetoconductance. (b) Electronic band structure in presence of in-plane magnetization. (c) The corresponding non-trivial Berry curvature with a hot-spot at the avoided crossing point.

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First principles study of NdNiO₃ – SrTiO₃ interfacesAlexander Lione, Nicholas Bristowe*Durham University*

ABO₃ Perovskites are a popular playground for both fundamental and applied research. They show a rich variety of structural, magnetic and orbitally ordered phase transitions. The properties of Perovskites may be further manipulated by interfacing them with one another. As a thin film deposited epitaxially on a substrate, the properties are altered due to lattice parameter mismatch resulting in biaxial strain, mismatch in structural or magnetic modes at the interface, quantum confinement due to film thickness, polar discontinuities due to formal or ferroelectric polarisations of either material, and more, depending on the choice of materials. For example, the discontinuity in the formal polarisation between nominally insulating LaAlO₃ and SrTiO₃ has been suggested to induce an emergent 2D electron gases (2DEG) at their interface [1].

We present an investigation into the interface between two perovskites SrTiO₃ and NdNiO₃, from first principles simulations based on density functional theory. Both materials exhibit important phase transitions with decreasing temperature, which can be manipulated by interfacial effects. SrTiO₃ is a quantum paraelectric in bulk [2], but may undergo a polar transition under biaxial strain [3]. NdNiO₃ is a metal at room temperature [4], but undergoes a metal-insulator transition (MIT) with reducing temperature. This transition may be controlled with biaxial strain [5], such that NdNiO₃ may even remain metallic at ground state. The FE and MI transitions are also heavily coupled to octahedral tilting [6,7], which is typically affected by mode decay across the interface, especially when the Perovskites have different tilt patterns, as is the case with SrTiO₃ (a⁰a⁰a⁻) [2] and NdNiO₃ (a⁻a⁻c⁺) [4]. Finally, we discuss the role of the polarisation discontinuity in these interfaces, in analogy to the LaAlO₃ - SrTiO₃ interface.

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[P1-46]

Electronic reconstruction and anomalous Hall effect in the $\text{LaAlO}_3/\text{SrRuO}_3$ heterostructure

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4d and 5d transition metal oxides are a promising class of materials for topological phases in the context of electron correlations. Recently, the ferromagnetic metal SrRuO_3 (SRO) grown on a SrTiO_3 (STO) (001) substrate has been reported to exhibit electronic reconstruction induced interfacial charge pinning accompanied by a topological transition of its electronic bands when capped with a LaAlO_3 (LAO) layer [1]. LAO is a polar oxide and the electronic reconstruction in a heterosystem of LAO/STO caused by the polar discontinuity at the interface is well known. For the LAO/SRO system a similar behavior is expected, and charge is thought to be accumulated at the very interface giving rise to strong inversion-symmetry breaking and hence change in the momentum-space topology [1]. By means of transport and photoemission experiments we find strong evidence for these statements. On one hand we show that the LAO capping drives the SRO, which turns insulating below 8 uc without capping, (deeper) into the metallic regime. Furthermore, we show not only the observation of signatures of an anomalous Hall effect (AHE) in 6uc SRO films capped with LAO, but also an inversion of the sign of the AHE, when the 4uc SRO is replaced by metallic 10uc of SRO indicating a topological phase transition. Moreover, we correlate these findings with hard and soft x-ray photoemission spectroscopy data, that show changes in the ruthenium electronic states, that can be explained in terms of correlated electrons.

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[P1-47]

Rule of common anions on interfacial charge transfer in oxide heterostructures studied by in-situ photoemission spectroscopy

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Electronic structures at interfaces composed of different materials are of great importance for the design of electronic devices and artificial functional materials. The band alignment at the interface is one of the fundamental parameters for designing electronic devices and artificial functional materials. However, there is no firmly established guideline for oxide heterostructures, limiting the functional design of oxide heterostructures. Here, we provide spectral evidence that the band diagram of oxide heterointerfaces is well described by the Zhong and Hansmann scheme based on the common anion rule [1,2]. By utilizing the elemental selectivity of Ti $2p$ - $3d$ resonant photoemission spectroscopy for the Ti $3d$ state near the Fermi level, we directly visualize the presence or absence of charge transfer from the overlayer films to SrTiO₃ in prototypical heterointerfaces of SrVO₃/SrTiO₃ and SrNbO₃/SrTiO₃[3]. It is found that the charge transfer occurs in SrNbO₃/SrTiO₃ but not in SrVO₃/SrTiO₃, as predicted by the Zhong and Hansmann scheme, indicating that the presence or absence, as well as the sign and amount, of interfacial charge transfer is predicted by this scheme. The detailed analysis reveals that the carrier density at the interface and the characteristic length scale are considerably different depending on the presence or absence of charge transfer. Our findings provide guidelines for designing and controlling the functionalities in oxide nanostructures.

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Large in-plane superconducting critical fields in highly confined LaAlO₃/KTaO₃(111) electron gas

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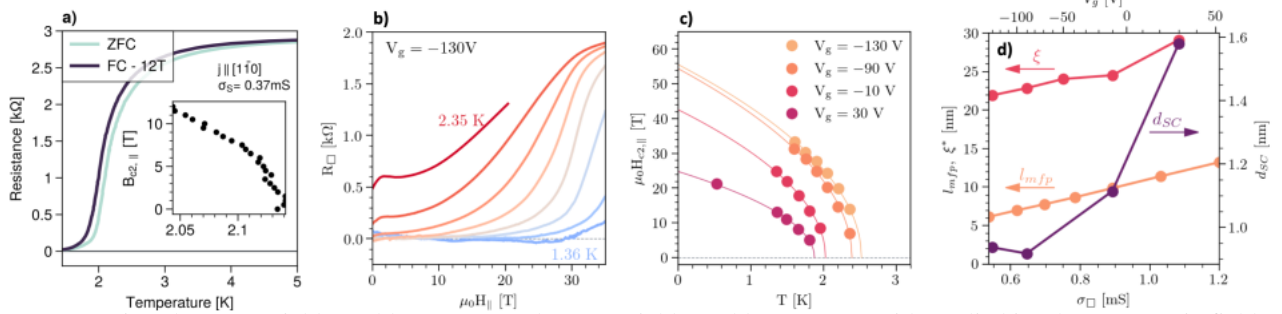
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Interfacial oxide superconductors can host non-conventional superconducting states due to their low dimensionality, low symmetry and strong spin orbit coupling (SOC). The discovery of superconductivity at the surface of KTaO₃(111) [1] sparked renewed interest in this research direction due to the uniqueness of the fully spin-orbit-coupled ground state and the non-coplanar spin texture of its Fermi surface.

In this study, we examine the superconducting critical fields of two-dimensional electron gases at the surface of KTaO₃(111) as a function of electrostatic back-gating. Our findings reveal in-plane critical fields of unprecedented magnitude in two-dimensional oxide interfaces, reaching up to 56T at 0K. By comparing in-plane and out-of-plane critical fields, we identify an extremely anisotropic superconductor with a thickness of less than 1 nm and up to a 12-fold violation of the Chandrasekhar-Clogston paramagnetic limit [2,3]. The analysis of the weak anti-localization indicates that the enhancement of the critical fields is due to an exceptionally thin superconducting layer and spin-orbit-scattering-suppressed paramagnetic susceptibility. Our research underscores the critical role of spin-orbit coupling and two-dimensional confinement in understanding superconductivity at oxide interfaces.



a) Comparison between Field-Cooldown (FC) and a Zero Field-Cooldown (ZFC) with applied in-plane magnetic field. Inset: the dependence of the in-plane critical field on temperature. b) Sheet resistance vs applied in-plane magnetic field for different temperatures. c) Measured in-plane critical field vs temperature for different values of back-gate voltage.

The solid line represents a fit to the 2D-Ginzburg-Landau model, d) Coherence length (ξ) and thickness of the superconducting layer (d_{SC}) compared to the normal state mean free path (l_{MFP}).

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Ferroelectric two-dimensional electron gases based on strain engineered SrTiO₃ thin films

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Two-dimensional electron gases (2DEGs) based on the quantum paraelectric SrTiO₃ (STO) display fascinating properties such as large electron mobilities, superconductivity and efficient spin-charge interconversion owing to their Rashba spin-orbit coupling [1-3]. However, such 2DEGs have almost exclusively been generated in STO single crystals, with the few attempts to replace crystals by heteroepitaxial STO thin films leading to low carrier mobilities. This is limiting the potential to integrate STO 2DEGs in future devices as well as the possibility to introduce additional functionalities specific to STO thin films, such as strain-induced ferroelectricity. Here, we employ oxide molecular beam epitaxy to grow high quality strain-engineered STO films on LSAT that are ferroelectric up to 170 K. We then generate a 2DEG by sputtering a thin Al layer and demonstrate an increase in both the low and room temperature mobilities by up to factor of four compared to earlier literature. Furthermore, through Raman spectroscopy and magneto-transport measurements, we show that the ferroelectric character is retained after 2DEG formation which is further confirmed by scanning transmission electron microscopy (STEM) at low temperature. Real space visualization of the 2DEG is obtained with monochromated electron energy loss spectroscopy at the Ti-L_{2,3} edge, evidencing Ti³⁺ in unit-cells near the interface. STEM-geometrical phase analysis at low temperatures (~ 110 K) indicates a c-axis gradient, possibly associated with the ferroelectric transition. These findings establish our samples as ferroelectric 2DEGs operational at temperatures significantly higher than those reported for Ca-doped STO substrates (~30 K) [4], paving the way for the development of ferroelectric 2DEGs capable of functioning at room temperature.

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Towards magnonic spin current injection at oxide interface-based two-dimensional electron gases

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The groundbreaking discovery [1] of a high mobility two-dimensional electron gas (2DEG) at the interface between two insulating oxides, namely, SrTiO₃ (STO) and LaAlO₃ (LAO) attracted enormous attention because of their intriguing physical properties *e.g.*, interfacial magnetism, gate-tunable Rashba spin-orbit coupling (SOC) and high spin-to-charge conversion efficiency. When a spin current is injected from a ferromagnet (FM) to the 2DEG perpendicular to the interface, an in-plane charge current in the 2DEG is induced via the inverse Edelstein effect (IEE).[2] The efficiency of the spin-to-charge current conversion via the IEE (λ_{IEE}) is proportional to the Rashba coefficient (α_R) and the mobility of the electrons (μ_e) in the 2DEG. For an efficient spin-charge conversion, it is essential to fabricate oxide-based heterostructures consisting of a FM film with low Gilbert damping (α_{FM}) and a 2DEG with high values of α_R and μ_e . While a large value of λ_{IEE} up to ~ 190 nm has been obtained in the 2DEG at the LaTiO₃/SrTiO₃ interface for $\mu_e \approx 10^3$ cm²/V.s at 2 K, [3] it was experimentally shown that a very high mobility 2DEG with $\mu_e > 10^5$ cm²/V.s at 2 K can be formed at the interface between spinel-structured γ -Al₂O₃ (GAO) and STO [4]. Therefore, the 2DEG at the GAO/STO interface could potentially support very large values of λ_{IEE} at low temperatures. However, spin-charge conversion in GAO/STO based 2DEG system remains unexplored.

It is known that La_{0.7}Sr_{0.3}MnO₃ (LSMO) is a half-metallic FM with high Curie temperature ($T_C \approx 350$ K) and robust spin polarization, with only ≈ 0.75 % lattice mismatch with STO, and has an ultra-low α_{FM} ($\approx 10^{-3}$ – 10^{-4}) [5]. Although spin-pumping effect has been used in most of the studies reported in the literature so far to generate magnonic spin currents in conducting magnetic materials (*e.g.*, LSMO, permalloy) followed by spin angular momentum transfer to the adjacent oxides-based 2DEG to determine the spin-to-charge conversion efficiency [6], it may induce parasitic effects in case of conducting FMs which lead to the generation of spurious signals that require additional efforts to disentangle it from the intrinsic signal. Therefore, for intrinsic magnonic spin current generation without the associated Joule heating losses and effective spin angular momentum transfer from the FM to the 2DEG an insulating ferromagnetic material is desired. We have identified the room temperature ferromagnetic insulator NiFe₂O₄ (NFO) as a potential spin current source to investigate the spin-charge conversion in the 2DEG at the GAO/STO interface, as (a) both NFO and GAO possess a cubic spinel structure with nearly 5% lattice mismatch and (b) NFO also has an ultra-low value of α_{FM} ($\approx 10^{-4}$) [7] just like LSMO. Using a reactive radio frequency (RF) sputtering setup, we demonstrate that an insulating ferromagnetic layer of NFO can be grown on the top of GAO/STO film without compromising the high mobility 2DEG at the GAO/STO interface. Secondly, we also show the preservation of electrical conductivity in a LAO/STO-based interface when stacked with a freestanding ferromagnetic LSMO film.

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Combined TOF-SIMS, STEM-EELS and magneto-transport study of the two-dimensional electron gas at the $\text{AlO}_x/\text{KTaO}_3$ (110) interface

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Oxide-based two-dimensional electron gases (2DEGs) have garnered significant interest due to their potential for novel physical properties. Particularly, 2DEGs formed in KTaO_3 are noteworthy for their crystal face-dependent superconductivity and large Rashba splitting, promising for oxide electronics devices. This study presents an in-depth analysis of the 2DEG at the $\text{AlO}_x/\text{KTaO}_3$ (110) interface using time-of-flight secondary ion mass spectrometry (TOF-SIMS), scanning transmission electron microscopy-electron energy loss spectroscopy (STEM-EELS), and magneto-transport techniques. We identify the O-depletion region's extent, characterized by variations in KTaO_3^- and KTaO^- cluster signals during TOF-SIMS measurements, ranging from 2 to 4 nm. This region's correlation with AlO_x thickness in conducting heterostructures impacts the electronic structure, carrier density, and mobility of the 2DEG. We discuss these findings' implications for developing advanced oxide electronics.

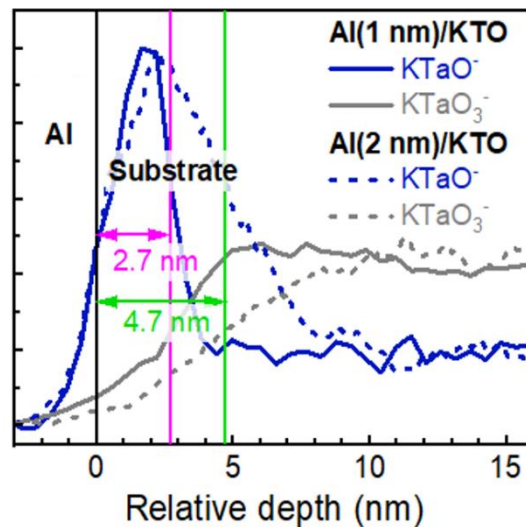


Figure 1. Visualization of the O-depleted region within the KTaO_3 substrate

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Giant Edelstein effect and non-linear transport in ferromagnetic Rashba 2D electron gases

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Strontium titanate SrTiO₃ (STO) two-dimensional electron gases (2DEG) have broken spatial inversion symmetry and possess a finite Rashba spin-orbit coupling, which endows the electronic bands with unique spin textures and couples the spin of mobile electrons to their momentum. This enables the interconversion of charge and spin currents through the direct and inverse Edelstein effects (EE/IEE), with record efficiencies at low temperature. In this work, we show that making these 2DEGs ferromagnetic enhances the conversion efficiency by nearly one order of magnitude. Starting from the experimental band structure of non-magnetic STO 2DEG [1], we mimic magnetic exchange coupling by introducing a Zeeman term in a tight-binding model. We then calculate the band structure and spin textures for increasing internal magnetic fields and compute the Edelstein effect using a semiclassical Boltzmann approach [2]. The conversion efficiency initially rises sharply with increasing magnetic field, peaks, and then decreases, due to the competition between exchange coupling and the effective Rashba interaction [3].

Experimentally, we study the 2DEG at the EuO/STO interface to imprint ferromagnetism in the gas. With XPS and SQUID characterization, we assess the 2DEG formation and the magnetization behavior of the ferromagnetic oxide layer. Through dc-transport measurements, we observe signatures of ferromagnetism, such as hysteretic in-plane magnetoresistance (MRIP) and the Anomalous Hall Effect (AHE) and show that the magnetism can be tuned with a back-gate voltage. Additionally, we conduct non-linear magnetotransport experiments to investigate the interplay between ferromagnetism and Rashba.

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[P1-53]

Synthesis and valence control of unconventional Ruddlesden-Popper nickelates by integrated electrochemistry approaches

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Novel nickelate materials have attracted considerable attention in the modern era. Synthesizing new nickelates is crucial for exploring their unique properties and underlying mechanisms [1]. The Ruddlesden-Popper (RP) phase offers an intriguing and tunable platform. In this study, we attempted to synthesize new nickelate materials by lithium-ion doping of La_2NiO_4 using electrochemical and solid-state reaction methods [2]. Our experiments revealed that both approaches effectively modulated the oxidation state of Ni, which significantly influences the magnetic properties of nickelates. These findings contribute to investigating novel nickelates and provide valuable insights into the relationship between chemical composition and material properties. Future work will focus on optimizing the electrochemical method and extending it to other materials, aiming to discover new nickelates with enhanced properties. This research advances the development of electrochemical synthesis techniques for nickelates and paves the way for the rational design of novel functional materials.

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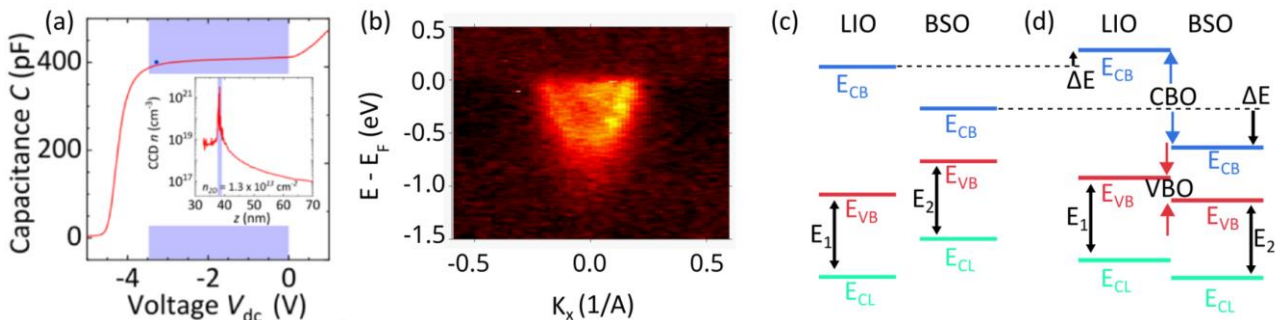
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Valence band offset determination beyond Kraut's method: Modelling and experiment for polar discontinuity doped BaSnO₃/LaInO₃ heterostructures

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Transparent semiconducting oxides (TSO's) are key players for new (opto-)electronic devices. For relevant device applications, both high electron mobilities and high charge carrier densities (CCDs) are required, bringing up the concept of TSO-based two-dimensional electron gases (2DEGs). In the search for more suitable TSOs, the wide-bandgap semiconductor BaSnO₃ (BSO) has been reported to provide the highest room-temperature (RT) electron mobilities - up to 320 cm²/Vs for La doped bulk material [1] - within the family of perovskites. For BaSnO₃/LaInO₃ (BSO/LIO) heterostructures, a predicted conduction band offset (CBO) of 1.6 eV confines a charge carrier density of up to 2*10¹⁴ cm⁻² in the quantum well at the BSO side [2].



(a) CV measurements revealing the presence of a charge accumulation layer at the BSO/LIO interface. Inset: Corresponding charge carrier density (CCD) profile as a function of layer thickness. (b) Band structure of the quantum well states at BSO/LIO interface showing two occupied subbands. (c) Sketch of conduction band (E_{CB}), Valence band (E_{VB}) and Core level energy (E_{CL}) of single BSO and LIO layers, and (d) of BSO/LIO heterostructure assuming flat bands.

Here, we investigate BSO/LIO heterostructures grown by plasma-assisted molecular beam epitaxy. The formation of the polar-discontinuity doped 2DEG at their interface is confirmed by capacitance-voltage (CV) [Fig. 1(a)] and Angle-resolved photo-electron spectroscopy [Fig. 1(b)]. Extensive hard x-ray photoelectron spectroscopy studies were performed to determine the valence band offset (VBO) beyond Kraut's method [3] – a method that assumes flat bands at the interface [Fig. 1(c) and 1(d)], a criterion that is not fulfilled at interfaces that host a 2DEG. X-ray energies ranging from 400 eV to 6.0 keV provide the depths resolution that not only give insights into band bending of the given heterostructure but also reveal the presence of the polarization field at the LIO/BSO interface. For determination of the VBO we developed a model that predicts XPS core-level shape and shift for given band profiles and thus related VBO correction for Kraut's method. Synchrotron-based XPS data are interpreted with this model to determine band bending and VBO at the BaSnO₃/LaInO₃ interface.

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Large nonlinear transverse conductivity and Berry curvature in KTaO_3 based two-dimensional electron gas

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Two-dimensional electron gas (2DEG) at oxide interfaces exhibits various exotic properties stemming from interfacial inversion and symmetry breaking. In this work [1], we report large nonlinear transverse conductivities in the $\text{LaAlO}_3/\text{KTaO}_3$ interface 2DEG under zero magnetic field. Skew scattering was identified as the dominant origin based on the cubic scaling of nonlinear transverse conductivity with linear longitudinal conductivity and 3-fold symmetry. Moreover, gate-tunable nonlinear transport with pronounced peak and dip was observed and reproduced by our theoretical calculation. These results indicate the presence of Berry curvature hotspots and thus a large Berry curvature triple at the oxide interface. Nonlinear transport offers a new pathway to probe the Berry curvature at oxide interfaces and facilitates new applications in oxide nonlinear electronics.

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[P1-56]

Growth controlled transport properties at two-dimensional LaAlO₃/KTaO₃ interfaces

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The simultaneous emergence of superconductivity and significant tunable spin-orbit coupling (SOC) in SrTiO₃ quasi-two-dimensional electron gases (q-2DEGs) presents a highly compelling system for the exploration of unconventional and topological superconducting phases. Intensive investigations have focused on q-2DEGs at the LaAlO₃/SrTiO₃ (LAO/STO) interface, where electrostatic gating facilitates the modulation of electron band filling within the t_{2g} manifold. This capability enables the electric field-induced modulation of the superconducting phase diagram below 300 mK and Rashba SOC. A groundbreaking recent discovery in this field is the observation of two-dimensional superconductivity in electron gases formed at the interface between (111)-oriented KTaO₃ and insulating LaAlO₃ overlayers. The superconducting transition temperature in this system, reaching up to 2.2 K, is approximately an order of magnitude higher than that observed in the LaAlO₃/SrTiO₃ interfaces. Furthermore, (110)-oriented KTaO₃ interfaces exhibit similar superconducting transitions, while the absence of superconductivity at (001)-oriented interfaces underscores the critical influence of band reconstruction. Additionally, a spontaneous in-plane transport anisotropy is detected prior to the onset of superconductivity, indicative of the emergence of a distinct stripe-like phase, which is dependent on electronic mobility. This warrants a systematic study of the effects of growth parameters on the fundamental properties.

In this study, we present a comprehensive investigation of electronic mobilities, carrier densities, and Rashba SOC in all three orientations of LaAlO₃/KTaO₃ interfaces as a function of growth temperature. We find an optimal growth temperature suitable for forming a homogeneous interface where anisotropic transport is minimal. We also find a systematic variation in both electronic mobilities and carrier densities. The Rashba SOC parameter, estimated from magneto-transport studies, reveals variations as a function of growth temperature.

2DHG or 2DEG? Spectroscopic insights into the substrate-assisted redox reaction of ultrathin Fe films on SrTiO₃

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The interface between two different oxide materials can exhibit emerging properties, on which novel oxide electronic devices can be based. For example, magnetism or ferroelectricity may be observed, or – as recently – the formation of either a two-dimensional electron (2DEG) or -hole gas (2DHG) in SrTiO₃ (STO) – apparently dependent on the oxidation state of a ferromagnetic metal overlayer [1]. In this context, the details of the substrate-assisted redox reaction are of major interest.

Ultrathin films of 3d transition metals (3dTM) iron, cobalt and their oxides with various thicknesses were deposited on TiO₂-terminated and Nb-doped STO substrates by molecular beam epitaxy (MBE). Using lab-based x-ray photoelectron spectroscopy (XPS) at $h\nu = 1486$ eV, the interface between 3d transition metals and the STO is investigated in situ.

The formation of oxygen vacancies in the STO substrate is revealed by the reduction Ti via spectral signatures of the chemical core level shifts [2]. In addition, rigid shifts in the binding energy of all core levels indicate a band alignment between film and substrate, suggesting either the emergence of a 2DEG or 2DHG at the STO interface.

On the 3dTM film side of the interface, small amounts of oxidized iron were found in the Fe 2p core level spectra, indicating diffusion of oxygen from STO across the interface into the iron thin film [3]. By varying the deposition temperature and 3dTM layer thickness, we further characterised the interface revealing a strong deposition temperature dependency of the redox reaction: For Fe deposition, we found a constant FeO layer thickness, while the redox reaction of similarly prepared cobalt thin films showed a redox behaviour with lesser extent.

These results give detailed insights into the formation of the interface between 3d transition metals and STO, which are necessary to design either 2DEGs or 2DHGs in novel 3dTM/STO interfaces.

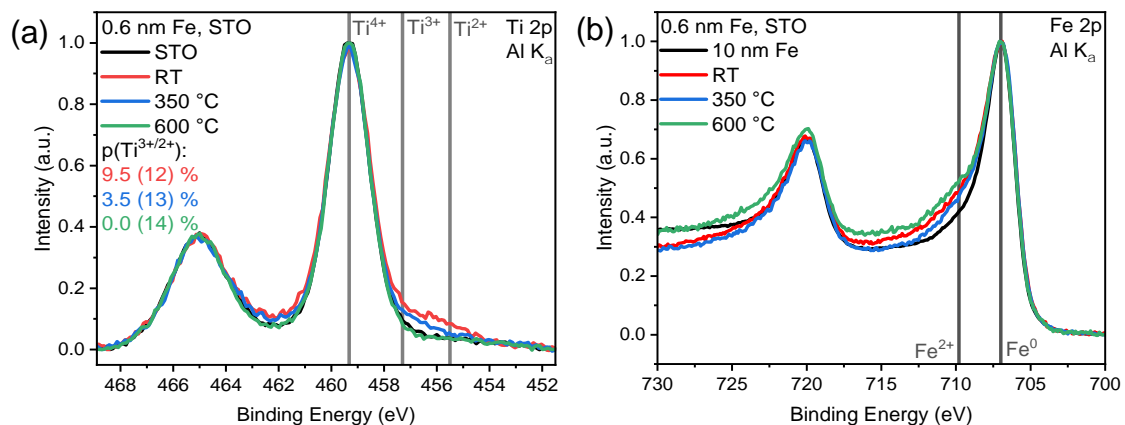


Fig. 1: XPS core level spectra Ti 2p (a) and Fe 2p (b) of a 0.6 nm ultrathin Fe film on STO:Nb substrate deposited at different temperatures. The Ti 2p core level reveals a reduction of Ti⁴⁺ and the Fe 2p core level indicates the oxidation Fe in the ultrathin film. This strong temperature-dependent redox reaction is caused by the transition of oxygen from the substrate across the interface inside the film.

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[P1-58]

High-temperature superconducting oxide without copper

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The 1987 Nobel Prize in Physics celebrated the discovery of superconducting ceramics (oxides) with transition temperature $T_c > 30$ K [1]. While Bednorz and Müller demonstrated the high-temperature superconductivity in the copper oxides, superconductivity with $T_c > 30$ K at ambient pressure has never been found in another ceramic. For the last four decades, it has been a puzzle for the question “Is high-temperature superconductivity ubiquitous among a broad class of oxides?” or “What makes Cu so special?” Nickel oxide was another promising candidate attempted by Bednorz and Müller. The prospects of ‘cuprate-like’ high- T_c in the nickel oxides have been under debate by theoretical and experimental reports [2-6]. In this talk, we will discuss our efforts with direct evidence to answer this question. Developed from our millikelvin penetration depth and 60 Tesla high-field magnetotransport studies, we propose a phenomenological theory which the superconducting states in the infinite-layer nickel oxides are highly tunable and the corresponding coupling strength can be maximized, therefore, leading to much higher T_c superconductivity [7, 8]. Evidence for such a manipulation and high- T_c superconductivity in a new oxide without copper under ambient pressure will be presented in this talk.

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Superconducting gap symmetry of nickel oxides

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Recently, superconductivity at 80 K was reported in a high-pressure induced tetragonal phase of a nickel oxide $\text{La}_3\text{Ni}_2\text{O}_{7+y}$ [1]. It is critical to understand whether nickel oxides share a similar pairing mechanism as of the copper oxides which superconduct at high-temperature under ambient pressure [2], especially noting the 3D character and lack of Ni-O hybridization in the electronic structures [3-4]. Determining the symmetry of the superconducting gap is the prerequisite for identifying the origin of the Cooper pairing in nickel oxides. Here, we performed millikelvin penetration depth measurement of the ambient-pressure nickel oxide superconductors $(\text{La,Ca})\text{NiO}_2$ and $(\text{Nd,Sr})\text{NiO}_2$ and theoretically derived the superfluid density from the pairing of nickel $3d_{x^2-y^2}$ orbital of various symmetries that satisfy its space group. We will discuss our calculations which show that a time-reversal-symmetry breaking order parameter of admixed d - wave symmetry: $(d+is)$ -wave is consistent with the experimental data. This contrast in the order parameters between nickelates and cuprates provides direct insights into the high-temperature pairing mechanisms [5-8].

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Structural and electronic properties of infinite layer NdNiO₂ and SmNiO₂

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Infinite layer nickelates have attracted considerable attention in recent times due to their intriguing electronic properties and potential for high-temperature superconductivity in this system isostructural and isoelectronic to the cuprates [1,2]. To date, superconducting nickel oxides have been stabilized in thin-film form by growing a perovskite rare-earth nickelate RNiO₃ (R = Nd, Pr, La) (113) - hole-doped with Sr or Ca, and topotactically reduced to the infinite-layer phase RNiO₂ (112) using CaH₂ or NaH as reducing agents [3,4].

In more recent studies, however, it was shown that a solid-state method employing the deposition of an ultra-thin aluminium layer on the perovskite film also allows the selective removal of the apical oxygen atoms, transforming the perovskite phase into the infinite layer one in a more reproducible way. The process is described by the following reaction: $2Al + 3NdNiO_3 \rightarrow Al_2O_3 + 3NdNiO_2$, and, in contrast to the chemical process described above, it should prevent the formation of secondary phases, impurities and residual hydrogen intercalation [5,6].

In this work, we use our experience in growing high quality 113 films and heterostructures [7], and utilize this procedure to reduce the 113 phase of both neodymium and samarium nickelate thin films to the 112 phase on different substrates. The samples involved in this study are grown by RF off-axis magnetron sputtering and the aluminium layer is subsequently sputtered in-situ on-axis.

A full structural characterization by means of X-ray diffraction (XRD) and scanning transmission electron microscopy (STEM), together with detailed transport measurements, will be presented.

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Systematic study of high energy electron irradiation induced disorder in superconducting nickelates to probe the order parameter symmetry

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Superconducting infinite-layer nickelates were experimentally realised two decades after their theoretical prediction [1], providing a platform to explore a close structural and electronic analogue to the high- T_c cuprates. This feat was achieved by combining epitaxial thin film synthesis with topochemical reduction to stabilise the required nickel electronic configuration [2]. Despite similarities in their respective phase diagrams [3], notable disparities in band structure and hybridisation have been identified [4-7].

Determining the pairing symmetry of these superconducting nickelate thin films has posed a significant challenge [8], partly due to sample geometry and surface degradation during chemical reduction processes [9]. Various techniques to investigate the superconducting order parameter symmetry, including studies of London penetration depth through mutual inductance [10] or tunnel diode oscillator method [11], single-particle tunnelling [12], photoemission spectroscopy [13-15], and thermal transport have thus far proved challenging to perform or interpret on these chemically reduced thin films.

We employ high-energy electron irradiation as a tool to controllably and systematically induce disorder into superconducting infinite-layer nickelate thin films and examine the impact of pair-breaking defects on the superconductivity. Our initial findings show the suppression of superconducting transition temperature and increase of normal state resistivity with induced disorder, shedding light on the nature of the gap symmetry and electronic landscape of the infinite-layer nickelates.

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Lack of hydrogen insertion in optimally synthesized infinite layer nickelates

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Infinite-layer nickelates are promising candidates for studying unconventional superconductivity because of their electronic and structural similarities with the cuprates [1]. Unconventional superconductivity in the nickelates was first realized in strontium-doped neodymium nickelate (Nd,Sr)NiO₂, which has since sparked a plethora of studies investigating Ni-based compounds [2,3]. The synthesis of infinite-layer nickelates, (RNiO₂, R = lanthanide), is a two-step process: first involving the growth of the perovskite precursor phase using pulsed laser deposition followed by the deintercalation of apical site oxygen atoms via topotactic reduction [4,5]. The topotactic structural transition is commonly achieved using calcium hydride (CaH₂) as a reducing agent [6,7]. It remains uncertain, however, whether the use of calcium hydride results in the insertion of hydrogen into the infinite layer structure [8–10]. To determine whether hydrogen is present in the infinite layer nickelates, we performed secondary ion mass spectroscopy (SIMS) measurements on optimally doped (Nd,Sr)NiO₂ thin films to quantify the hydrogen content resulting from calcium hydride reduction. We explore whether hydrogen plays a critical role for superconductivity in nickelates, and whether the presence of hydrogen is a result of non-crystallinity in from non-optimally synthesized thin films.

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Towards an alternative approach to achieve superconducting infinite-layer nickelate thin films

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After decades of research, a cuprate analog displaying superconductivity was finally found in the nickelate family. Superconductivity was reported in thin films of hole-doped infinite-layer nickelates ($\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$, x =dopant concentration) showing a $T_C \approx 10\text{-}15$ K [1]. Obtaining this superconducting phase is challenging since it requires the selective removal of all apical oxygens from the perovskite parent phase, which is usually achieved by an intricate topotactic reduction process using CaH_2 as the reducing agent. This method is highly impractical and prone to irreproducibility issues, which is a major obstacle in the progress of this new field of research. Therefore, innovative approaches for achieving superconducting infinite-layer nickelates in a easy and reliable manner are now needed.

In this work, we use in-situ sputtering to deposit an aluminum overlayer that acts as an oxygen scavenger to reduce the parent perovskite phase and attain superconducting infinite-layer thin films. We will present the whole optimization process of the Al deposition and the key parameters for successfully stabilizing the superconducting infinite-layer phase in thin films of different thicknesses by this alternative method. Our findings could help in improving sample reproducibility and crystallinity, besides providing an easier and more accessible method to obtain infinite-layer samples that may also be compatible with the use of surface sensitive techniques.

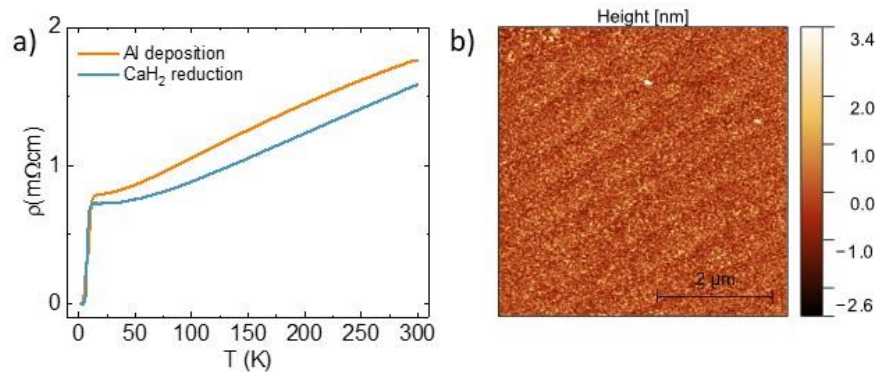


Figure 1. a) Temperature dependence of resistivity for superconducting $\text{Pr}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ infinite-layer thin films reduced by using CaH_2 (blue) and Al deposition (orange) methods. b) Atomic Force Microscopy (AFM) image of the surface of an infinite-layer sample obtained after Al sputtering deposition.

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Mechanism of reversible phase changes during hydrogenation of nickelate thin films

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Rare-earth nickelates $R\text{NiO}_3$ with $R=\text{La-Lu}$ are an interesting material system where a metal-to-insulator transition can be induced by hydrogen. Since the nickelates are sensitive to hydrogen, stable in harsh environments, compatible with conductive aqueous media and their resistance change controllable by electric field and reversible, they have been suggested for a range of applications: electric field sensors in salt water, bio sensing, electrolyte for solid oxide fuel cells, memory device, and neuromorphic computing [1-5]. Motivated by the many prospective applications this work focuses on understanding the underlying mechanism. We report on our experiments on hydrogenation of LaNiO_3 thin films capped with a catalytic $\text{Pd}_{0.6}\text{Au}_{0.4}$ layer and exposed to hydrogen gas, combining neutron reflectometry, x-ray spectroscopy and transport measurements. In contrast to previous studies, we observe not only a transition from the metallic to the insulating phase, but also a transition to another metallic phase with sufficient gas exposure. Additionally the hydrogenated state is found to be metastable and decays as soon as the surrounding hydrogen is removed, with the film subsequently becoming semiconducting. As indicated in the schematic annealing in air is required to restore the initial state closing the cycle. Our results reveal that the changes in electronic properties are linked not only to hydrogen incorporation, but also to a concomitant loss of oxygen. Thus, in addition to demonstrating the interesting possibility of hydrogen doping, our study underlines the importance of the oxygen stoichiometry in the nickelates.

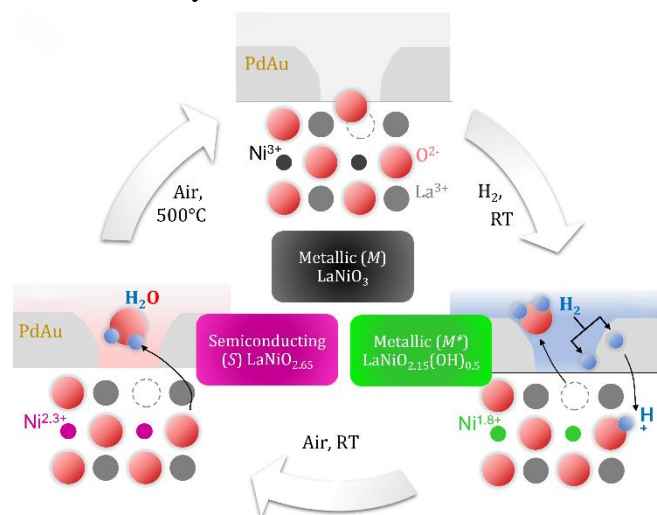


Figure 2: Schematic of the reaction cycle of a LaNiO_3 layer after exposure to hydrogen gas, air at room temperature (RT) and air at 500°C

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[P1-65]

Synthesis and Electronic Structure of Unconventional Nickelate Thin Films on Orthorhombic NdGaO₃ Substrates

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The discovery of nickelate superconductors has engendered reviving interest, and the nickelate family continues to grow [1-4]. Regarding the infinite-layer nickelates that possess a $3d^9$ electron configuration analogous to that of cuprates, many questions remain unresolved, for which synthesis of high-crystallinity infinite-layer thin films as well as novel phases present significant importance. Here, we report the synthesis of a high-quality NdNiO₂ on an orthorhombic NdGaO₃ substrate by RF magnetron sputtering and a low-temperature topotactic reduction using NaH. Nonlinear Hall effect and signatures of superconductivity were observed in these films. X-ray scattering spectroscopy and first-principles calculations revealed a large orbital polarization of the $d_{x^2-y^2}$ bands, and a reduced hybridization of the Nd $5d$ and Ni $3d$ orbitals. Furthermore, we also show the synthesis of an intermediate meta-stable samarium nickelate, Sm₉Ni₉O₂₂, between the perovskite and the infinite-layer phases [5]. This phase, formed by ordered apical oxygen vacancies in the intricate planes of $\{303\}_{pc}$, provides an essential materials paradigm, in which the square planar (Ni¹⁺), pyramidal (Ni²⁺), and octahedral (Ni³⁺) Ni sites co-exist. These two examples illustrate that topotactic reductions offer unparalleled opportunities to effectively prepare thermodynamically meta-stable nickelates, with the potential of finding additional superconductors and/or other exotic new phases.

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[P1-66]

Real-time monitoring of topotactic reduction of nickelate from perovskite to infinite layer phase

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The discovery of superconductivity in the infinite layer phase of hole doped nickelate has marked the beginning of a new era in nickelate superconductivity [1]. The NiO₄ square planar structure in the infinite layer leads to an Ni⁺ d⁹ configuration, which is isoelectronic to Cu²⁺ and analogous to the high T_c layered cuprates. Since the initial report [2], reproducibly producing good quality samples has been challenging. We present a method to monitor the topotactic reduction of perovskite to the infinite layer nickelate phase using CaH₂ in real time. We first discuss the evolution of resistivity as the perovskite transitions to infinite layer phase in samples with and without STO capping. We then provide further insight into the reduction process through structural characterizations (XRD and TEM) at different stages of this resistivity evolution. Our findings show that the gradual reduction in the c-axis starts closer to the substrate interface, as the oxygen deintercalation requires diffusion through the outer film layers. Our characterizations highlight the crucial role of STO capping in stabilizing the infinite layer nickelate phase and preventing decomposition on the top layer of the film. However, STEM characterization reveals the presence of a transition layer (~ 2 u.c) near the top interface with STO capping. A summary of all reduction run identifies an ideal temperature window for reduction of Sr-doped NdNiO₃ system, with 310°C being the thermodynamic sweet spot where the infinite layer can be achieved over a wide range of end-pressure vis a vi CaH₂ powder quantity. Using these conclusions, we achieved record values of T_{c,0}, T_{c,onset}, and Residual Resistivity Ratio (RRR) of 13.8 K, 19.8 K, and greater than 6, respectively, for a ~2nm STO-capped Nd_{0.8}Sr_{0.2}NiO₃ sample on an STO substrate.

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Synthesis and characterization of lanthanides nickelates thin films

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In 2019 [1], a new family of superconducting thin films was discovered: nickelates. That property is report in lanthanide doped by strontium nickel infinite layers (Pr, Nd, La) [2]. Infinite layers are synthesized by a topotactic reduction with calcium hybride of the perovskite phase [3]. The theoretical structure [1] with in plane NiO_2 could explain the superconducting property. The aim of this project is to understand the mechanism of the superconductivity in those compounds and study the structure to link it with physical properties.

First step is to synthesize high qualities thin films of doped and undoped perovskite of $\text{Pr}_{1-x}\text{Sr}_x\text{NiO}_3$ and $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3$ by pulsed laser deposition and reduce them in infinite layer phases $\text{Pr}_{1-x}\text{Sr}_x\text{NiO}_2$ and $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$. The X-ray fluorescence highlighted a deficit of nickel in our thin film and it could be link to transport properties (resistivity).

Infinite layers of $\text{Pr}_{1-x}\text{Sr}_x\text{NiO}_2$ are synthesize and preliminaries measurements show a strong impact of the strain in the homogeneity of thin film. A study of transport properties of infinite layers of $\text{Pr}_{1-x}\text{Sr}_x\text{NiO}_2$ evince more strain there is in the structure more the reduction process is inhomogeneous.

Hall effect measurements are performed to calculate the density of charge carriers and the mobility of them in all compositions (NNO, NSNO, PNO, PSNO) before reduction. The realization of that measure on our thin film on different substrates (SrTiO_3 and LSAT) allow us to understand the real impact of the strain and the possible influence of defects generated during the growth or reduction process. This study of charge carriers' density is compared with theoretical value in YBaCuO which is a familiar superconducting material.

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Probing the magnetism of infinite-layer nickelates with quantum magnetometry and X-ray dichroism

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The correlation between antiferromagnetism (AFM) and superconductivity is considered key to understanding the mechanism behind oxide high-temperature superconductivity in cuprates. [1] However, in infinite-layer (IL) nickelates, which are analogous to cuprates, the long-range AFM order is absent. [2,3] Instead, spin-glass behavior and antiferromagnetic spin fluctuations have been reported in these materials. [4,5] Despite numerous previous efforts, current experimental studies have not clarified the magnetic nature of the Ni and rare earth elements. Hence, it is vital to perform element-resolved detection of magnetism in nickelates. Additionally, Ni impurities (e.g., NiO_x clusters) influence magnetic-scanning-imaging results, adding a ferromagnetic signal that complicates the probing the ground magnetic states of IL nickelates. [6]

To address these issues, we have optimized the epitaxial growth and topotactic reduction process to prevent the formation of NiO_x clusters in our samples. We then utilized quantum magnetometry to conduct nanoscale magnetic imaging on a superconducting Nd_{0.8}Sr_{0.2}NiO₂/SrTiO₃ sample, observing distinct magnetic domains of ~100 nm. Additionally, we employed X-ray magnetic circular dichroism (XMCD), at the Ni-L_{2,3} and Nd-M_{4,5} edges, to confirm the elemental origin of magnetism in Nd_{1-x}Sr_xNiO₂ (x= 0~0.2) film samples, observing clear distinctions between Ni and Nd contributions. Intriguing temperature and Sr-doping dependence of the Ni-L edge XMCD were also found. These results demonstrate the unique magnetic nature of nickelates compared to cuprates, suggesting IL nickelates as a promising platform for investigating unconventional superconductivity.

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[P2-1]

The role of oxygen octahedra connectivity in orthorhombic perovskite heterostructures

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Controlling the atomic structure in epitaxial transition metal perovskite thin films is an effective way to tune their properties or to achieve new functionalities. This is linked to the intimate coupling of the electronic and structural degrees of freedom in this family of materials. For instance, the rotation angle of the oxygen octahedra is directly linked to the electronic bandwidth in nickelates or to the orbital ordering temperature in vanadates. Another example is a predicted novel ferroelectric state due to a tailored coupling of rotations in epitaxial heterostructures of different symmetries [1].

Achieving such a control requires an understanding of the mechanisms that determine the atomic structure - and may compete - in an epitaxially strained structure. In this work, we investigate orthorhombic perovskite films grown by pulsed laser deposition on orthorhombic substrates [2]. We show that there is a competition between oxygen octahedra connectivity and macroscopic strain leading to an “intermediate layer” and a “switching plane” at the interface that allow a re-orientation of the orthorhombic unit cell [3]. The detailed atomic structure of these oxide material combinations is probed using X-ray diffraction and scanning transmission electron microscopy.

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Interfacial charge-transfer in 3d/5d oxide heterostructuresD. Fuchs¹, A. K. Jaiswal¹, F. Wilhelm², A. Rogalev², and M. Le Tacon¹¹ *Karlsruhe Institute of Technology, Institute for Quantum Materials and Technologies,
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The synthesis of artificial oxide heterostructures and superlattices has gained increasing interest during the last decade due to the ability of epitaxial growth with atomic precision. The combination of different complex oxides not only results in the possibility of tuning the intriguing phenomena of the bulk constituents via lattice strain and symmetry change but also often generates new exotic phases at the interface of the correlated electron systems. Interfacial charge-transfer (ICT) between two different oxide layers, such as in the seminal experiments on LaAlO₃/SrTiO₃ heterostructures, seems to be most prominent for these observations. Besides the combination of 3d perovskites, which generally display strong electron correlation, heterostructures consisting of 5d perovskites additionally display distinct spin-orbit coupling, which might be useful for *e. g.*, spin manipulation in spintronic devices. In the following, we have studied systematically the ICT in 5d SrIrO₃ (I) heterostructures in combination with the 3d perovskites LaMnO₃ (M), LaFeO₃ (F), LaCoO₃ (C) and NdNiO₃ (N). High quality heterostructures [I_{*i*}X_{*i*}]_{*m*} (X = M, F, C, N) with a layer thickness of *i* monolayers each and a periodicity of *m* were prepared on (001) oriented SrTiO₃ substrates by pulsed laser deposition. The ICT was studied by measurements of the Hall resistance and x-ray absorption spectroscopy at the Ir *L*_{3,2} edge. For all the samples an electron transfer from the 5d⁵ I to the neighbored 3d^{*m*} layer X was observed. The ICT was found to be confined to the first layer with respect to the interface and systematically increases with increasing *n* up to *n* = 6 amounting to $\Delta n \approx 0.35$ electrons. For *n* = 7 (X = N) ICT reduces again. The increase of Δn with *n* can be well understood assuming the alignment of oxygen states at the interface due to the continuity of the common oxygen matrix by the corner sharing I and X octahedra [1], whereas the reduced ICT for X = N is likely explained by hybridization effects in the N layer, lowering Δn significantly with respect to LaNiO₃[2].

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Tuning the interlayer coupling in $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.95}\text{Ru}_{0.05}\text{O}_3/\text{LaNiO}_3$ multilayers with perpendicular magnetic anisotropy

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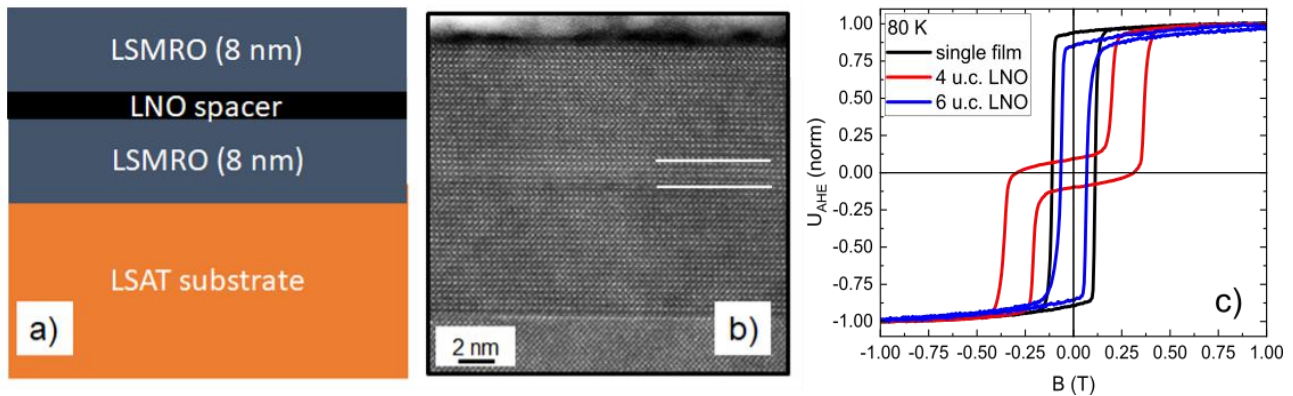
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For ferromagnetic oxide epitaxial multilayers the magnetic properties and the interlayer coupling are determined by manifold effects. Apart from the contribution of the interlayer exchange coupling, strain and interfacial effects, such as structural reconstructions or charge transfer, can play important roles and the resulting magnetic properties are thus a complex outcome. Here the interlayer coupling of ferromagnetic $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.95}\text{Ru}_{0.05}\text{O}_3$ (LSMRO) layers was investigated, when they are separated by epitaxial spacers of paramagnetic metallic LaNiO_3 (LNO), for stacks with perpendicular magnetic anisotropy. The thickness of the spacer was varied and it was found that the coupling between two LSMRO layers changes between antiferromagnetic (for a 4 unit cells thick LNO spacer) and ferromagnetic (for a 6 unit cells thick LNO spacer). For multilayers with five LSMRO layers and four unit cell thick LNO spacers, the antiferromagnetic coupling was preserved, but the effective magnetic anisotropy changed, canting the magnetization more toward in-plane. This behavior of the multilayer was confirmed by X-ray magnetic circular dichroism (XMCD) investigations at the Mn and Ni L_3 edges. XMCD indicated that the 4 unit cells thick LNO spacers of the multilayer become magnetically ordered, following closely the magnetization of the LSMRO layers. Future work will be devoted to LSMRO/LNO multilayers, their dependence on the thickness and number of LSMRO layers, and the study of the magnetic domains in antiferromagnetically coupled multilayers.



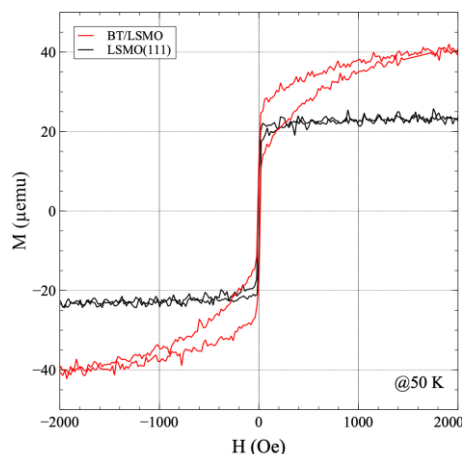
a) Schematic drawing of trilayer samples. b) High magnification HAAD-STEM image of a trilayer with 4 u.c LNO spacer. c) Anomalous Hall voltage at 80 K for a single LSMRO film (black), trilayer with 4 u.c. LNO spacer (red), and trilayer with 6 u.c. LNO spacer (blue).

Growth control of magnetic perovskite/2D topological insulator heterostructures - towards tailoring interfacial magnetic interactions

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Quantum materials gain an increasing interest in the field of electronics research. One exciting opportunity is heterostructures of magnetic oxides (MO) and topological insulators (TI) exhibiting unique combined properties and emergent interfacial properties. Recent studies on the topological insulating chalcogenide systems, forming heterostructures with magnetic compounds reveal that such systems exhibit emergent interface ferromagnetism, quantum anomalous Hall effect, enhancement of magnetic order, and ordering temperatures and topological surface states extending into the magnetic material. For the materials possessing inherently different crystal structures, an important first step is to ensure high-quality interfaces and the proper connectivity. In this work, we focus on structural engineering of TI Bi_2Te_3 (BT) grown on (001) & (111) oriented magnetic perovskites, as well as the importance of the interface analysis for understanding the origins of the emergent magnetic properties. The aim of the research is to establish a precise BT growth control towards a thorough understanding of magnetic interactions at magnetic perovskite-2D topological insulator interfaces. BT thin films are deposited by pulsed laser deposition (PLD) directly onto (001) and (111)-oriented SrTiO_3 (STO), $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) and LaFeO_3 (LFO). In structural analysis with SEM/AFM, XRD and TEM, we show that BT films can be grown by PLD on high lattice mismatch substrates, yielding high quality plane stacking and surface morphology. We employ Raman spectroscopy to confirm the proper BT stoichiometry. Vibrating sample magnetometry (VSM) is used to investigate the effect on macroscopic magnetism of ferromagnetic LSMO. The magnetization analysis indicates a possible magnetic proximity effect in the BT/LSMO(111) system. By utilizing different substrate orientations, as well as magnetic characteristics, heterostructure interactions can be tuned. This tunability can be used to further probe systematically how the oxide spin axis couples to the van der Waals material, and what is the effect on the topological insulator properties.



VSM data acquired for ferromagnetic LSMO(111) (black) and BT/LSMO (red). The magnetization characteristics, as well as the saturation is changed upon depositing the BT film.

Superlattice domain engineering by pulsed laser deposition

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Antiferromagnetic spintronics are considered to be a promising candidate for mitigating current rapid approach to semiconductor limitations. Antiferromagnets benefits from excellent robustness and terahertz frequency unlike its commonly used ferromagnet counterpart, while retaining the desired property to use electron spin as information carrier. Antiferromagnetic transition metal perovskites are of great interest in this regard for their highly dependent structure – functionality relation, especially in form of thin films for their substrate enabled strain engineering. In this work, we present our work in engineering of heterostructure superlattice in the (111)_{pc} orientation on anisotropic strained substrate, in which displays orientation-selected relaxation and its related magnetic behaviour.

(La_{0.7}Sr_{0.3}MnO₃/LaFeO₃)_{x4}/DyScO₃(101)_o superlattice is deposited by pulsed laser deposition with standard HF pre-treated DSO substrate for step-and-terrace finish. The as deposited super structure displayed uniform step-and-terrace as seen by atomic force microscope, that is fully strain from reciprocal space map (RSM) in Q_x= (-1 0 1)_o orientation. However, the super structure is seen to be relaxed in Q_x=(0 1 0)_o in RSM. Further investigation by 4D-STEM techniques revealed formation of structural domains with vertical domain walls separating two domains. The domains can be interpreted as double of unit cell which gives a second order Laue zone diffraction contrast in STEM. Further on, x-ray magnetic dichroism data from Advanced Light Source showing in-plane antiferromagnetic signature in ferromagnet hard axis and will be further discussed.

Oscillatory conduction behavior in an all-epitaxial (La_{2/3}Sr_{1/3})MnO₃/SrTiO₃/Nb:SrTiO₃ tunneling heterostructure

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Forming resonant levels using electron confinement has been demonstrated for perovskite-oxide heterostructures. Negative differential resistance with a single peak was observed in the conductivity-voltage curve of some heterostructures [1,2], and more recent works on quantum well (QW) structures reported resonant tunneling with multiple conductivity peaks [3,4]. However, in those heterostructures, elaborate techniques, such as utilizing multiple δ -doped layers and ferroelectric domain walls, were needed to induce conduction oscillations. In this work, we demonstrate clear oscillations of electrical conductivity in a simple oxide-based heterostructure with a SrTiO₃ (STO) tunnel barrier between La_{2/3}Sr_{1/3}MnO₃ (LSMO) and Nb:STO electrodes.

We epitaxially grew a heterostructure composed of 12-nm-thick LSMO / 10-nm-thick STO on a Nb-doped STO (001) substrate with a Nb concentration of 0.5 wt% using molecular beam epitaxy (MBE) [Fig. 1(a)]. After making mesa structures using electron beam lithography and Ar milling etching, the sample was annealed under pure oxygen at atmospheric pressure to remove oxygen vacancies. In this heterostructure, differential conductivity oscillations with multiple conduction peaks are visible in the tunneling transport measurements [Fig. 1(b,c)]. We do not see any hysteresis in the current-voltage curves [Fig. 1(d)], unlike the previous studies, which attributed the observed resonant electronic states to impurities or oxygen vacancy [1,2]. The tunnel resistance moderately increases with decreasing temperature, indicating that a leakage current is *not* significant in the STO barrier layer. The observed oscillations probably arise from the resonant states of electrons confined in the LSMO layer between the STO barrier and the sample surface, demonstrating that the simple oxide structure obtained by MBE is capable of oscillatory conduction.

This work was partly supported by Grants-in-Aid for Scientific Research, CREST and ERATO of JST, and the Spintronics Research Network of Japan (Spin-RNJ).

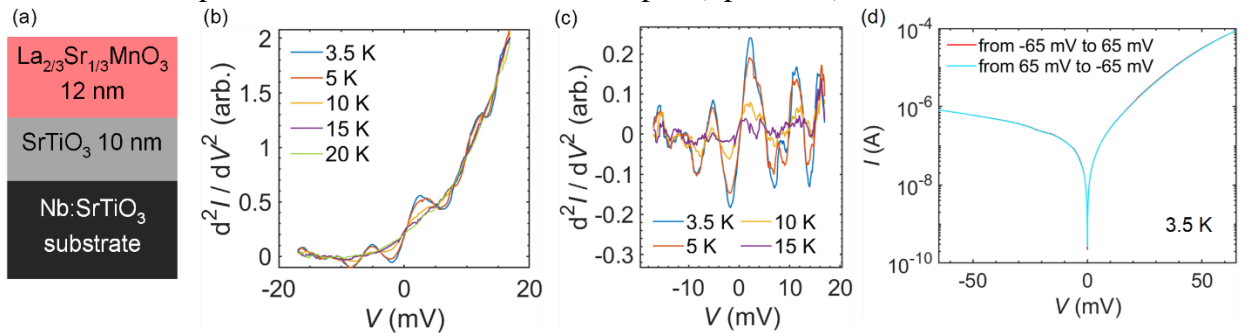


Fig. 1 (a) Schematic cross-sectional illustration of the LSMO/STO/Nb:STO heterostructure used in this study. (b) Derivative of the tunneling conductance obtained at various temperatures. (c) Derivative of the tunneling conductance at 3.5 K, 5 K, 10 K, and 15 K, after subtracting the data obtained at 20 K. (d) Current-voltage curve of the heterostructure when the bias voltage is scanned from -65 mV to 65 mV (red) or from 65 mV to -65 mV (cyan).

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Engineering the magnetic transition temperatures and the rare earth exchange interaction in oxide heterostructures

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The properties of functional oxide heterostructures are profoundly affected by the physics governing their interfaces. Modern deposition techniques allow for precise engineering of the interface physics through the growth of atomically precise heterostructures, granting meticulous control over the electronic, magnetic, and structural characteristics. In this study we show how to manipulate the transition temperatures and the exchange interaction in oxide superlattices using their periodicity. This is exemplified in superlattices of the insulating ferromagnetic double perovskites $\text{La}_2\text{NiMnO}_6$ (LNMO) and $\text{Nd}_2\text{NiMnO}_6$ (NNMO). First, a magnetization vs. periodicity phase diagram is shown, demonstrating that the respective para- to ferromagnetic transitions of LNMO and NNMO converge as the superlattice periodicity is reduced (See Fig. 1a). Our data is fitted to a simple Landau model that suggests a propagation of the magnetic order parameter across the superlattice interfaces. Second, we disentangled the magnetic system into a robust ferromagnetic Ni/Mn sublattice and a paramagnetic Nd sublattice with a low-field reversal of the Nd magnetic moments, showing how the Nd-Ni/Mn exchange coupling can be uniquely tuned at the interfaces (See Fig. 1b).

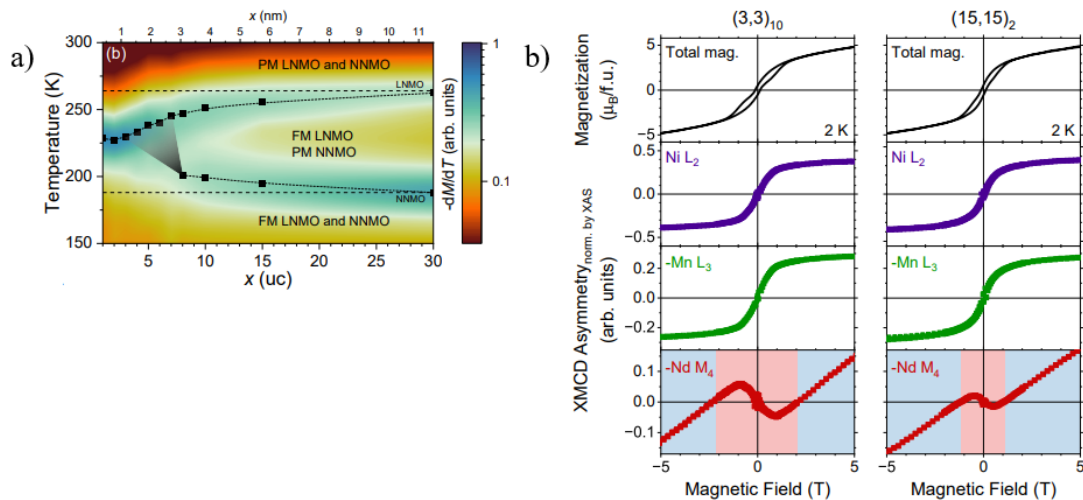


Figure 1: a) Phase diagram showing dM/dT as a superlattice period x and temperature function. b) $M(H)$ characterization at 2 K for a $(3, 3)_{10}$ and a $(15, 15)_2$ sample. The top row shows the total magnetization measured by SQUID. The remaining rows show the XMCD asymmetry measured at the indicated absorption edges.

Large spin-valve effect in a $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ -based spin-MOSFET with two easy-magnetization axes induced by inserting a LaMnO_3 buffer layer

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Exploring efficient spin-dependent transport is key to realizing next-generation ultra-low-power information processing devices. In particular, a spin-MOSFET is expected to be a new nonvolatile device that can significantly reduce the power consumption in logic circuitry. Previously, we obtained an extremely high magnetoresistance (MR) ratio of up to 140% at 3 K in a spin-MOSFET device based on single-crystal $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO) [1]. This MR ratio is 10–100 times larger than those reported in semiconductor-based spin-MOSFETs. In this device, we formed a nanometer-sized semiconducting channel by irradiating Ar ions to this region [2]. We analyzed the I - V characteristics and the back-gate modulation, concluding that the conduction occurs near the interface between the Ar-irradiated LSMO and a SrTiO_3 (STO) substrate. In this presentation, we show the spin-dependent transport properties in a new device in which we inserted a ferromagnetic LaMnO_3 (LMO) layer between the LSMO layer and the STO substrate. LMO has an electronic structure similar to Ar-irradiated LSMO. Thus, we can expect the channel properties to be more controllable.

We grew an LSMO (12 nm)/ LMO (6 nm) heterostructure on an STO (001) substrate using molecular beam epitaxy (MBE) [Fig. 1(a)]. Our magnetization measurements showed that the Curie temperature of the thin LMO layer in our device is 110 K. We prepared a spin MOSFET device using the same procedure as that described in Ref. [1] and measured the spin-dependent transport by varying an in-plane applied magnetic field. Fig. 1(b) and (c) show a representative MR curve and the plot of MR curves relative to the applied magnetic-field direction θ measured from the [100] direction, respectively. We observed large MR ratios of up to 10%. The color map shows anisotropic peaks of the MR ratio at $\theta = 60^\circ, 105^\circ, 240^\circ,$ and 285° [red circles in Fig. 1 (c)]. This feature may be explained by a combination of the easy magnetization axes of LSMO and LMO, which are [110] and [010], respectively. Fitting Simmons' equation to the experimental I - V curve yields that the barrier height formed near the LSMO/LMO interface is 81 meV, the length of the channel region is 4.4 nm, and its thickness is 1.2 nm. The obtained barrier height that is larger than that obtained in Ref. [1] (55.5 meV) suggests that the barrier height is increased by inserting the LMO layer [3].

This work was partly supported by Grants-in-Aid for Scientific Research, CREST and ERATO of JST, and the Spintronics Research Network of Japan (Spin-RNJ).

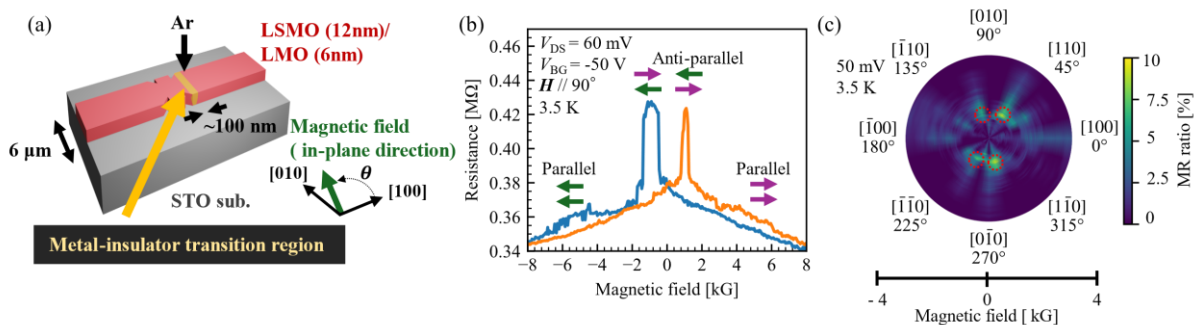


Fig. 1 (a) LSMO/LMO electrodes (red) separated by a tunnel barrier (yellow). (b) Magnetic-field dependence of the resistance. (c) Color map of the MR ratio of our device with respect to the applied magnetic field direction θ . The angle θ of the applied magnetic field is defined in Fig. 1 (a).

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***Operando* transmission soft x-ray absorption spectroscopy of Li-ion battery structure by x-ray excited optical luminescence**

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Soft x-ray absorption spectroscopy (XAS) is an excellent method for the direct observation of electronic states with element selectivity. It allows the observation of valence modulations due to chemical reactions as well as modulations of crystal structure from the ligand field. Therefore, soft XAS is expected to be used to elucidate not only the basic material properties but also the mechanisms of the device operation in the fields of material and energy sciences. However, because it is necessary to thin the thickness of the sample down to several hundred nm due to the short probing depth of soft x-ray, there are barriers to the *operando* observation of changes in electronic states of multilayered device structure under the operation using soft XAS measurements. Thus, an unconventional soft x-ray spectroscopy technique, where the thinning of the sample is not required, and we can directly measure the soft XAS on the actual device structure, is indispensable for *operando* observation of changes of electronic states under the operation.

We proposed an unconventional *operando* soft XAS method that can measure changes in electronic states of device structure under the operation, which is called “transmission x-ray absorption spectroscopy by x-ray excited optical luminescence (Tr-XAS by XEOL)”. In this method, a thin film and a device structure are prepared on a fluorescent substrate that emits visible light fluorescence (XEOL) by soft x-ray irradiation. XAS spectra are obtained by measuring XEOL generated from a fluorescent substrate by the transmitted soft x-ray through a thin film and a device [1]. Although this method is a kind of Tr-XAS, it does not require sample thinning, such as removal of the substrate, and soft XAS measurements can be performed on thin films with substrates and multilayered device structures. In addition, the signal intensity of Tr-XAS by XEOL is expected to be several orders of magnitude higher than that of the fluorescence XAS, allowing for faster scanning during the *operando* measurements.

In this presentation, we introduce the results of the developed *operando* Tr-XAS by XEOL of the thin-film all-solid-state Li-ion battery structure fabricated on an Al₂O₃ fluorescent substrate. The continuous valence changes of Co ions between Co³⁺ and Co⁴⁺ in the cathode electrode (LiCoO₂) following the charge-discharge reactions have been clearly observed from Tr-XAS spectra.

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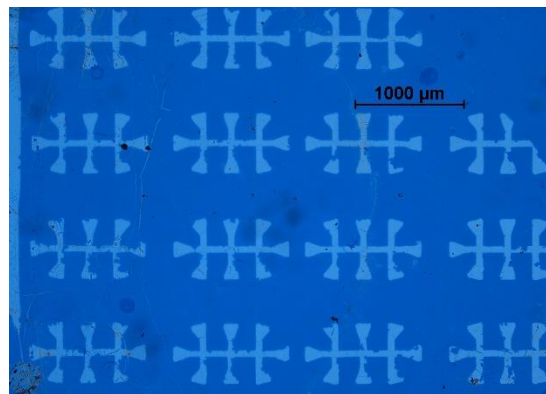
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Custom-shaped freestanding conducting oxide membranes on demand

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Creating freestanding oxide membranes with shape control opens the possibility to study a highly desirable range of feature sizes for future experiments and devices. One of the main challenges in patterning freestanding membranes is the tedious process required to synthesize the films, release and transfer them [1], and then shape them into a desired design. The question is whether we can achieve the desired shape in a single step, thereby eliminating numerous intermediate fabrication stages and allow the study of devices where the twist angle with respect to the new substrate can be varied systematically. Here, we present a simple and straightforward procedure to produce patterned freestanding membranes, which requires minimal laborious and time-consuming fabrication steps. Deposition of thin films is carried through shadow masks, which are placed between the material source (in this case, a PLD target) and the sacrificial layer on which the material is deposited. By doing this, a thin layer of material is deposited and structured simultaneously. The patterned structure is then transferred on a substrate of choice, in this case on SiO₂. The resolution of a shadow mask depends on two key parameters: the size of its features and the thickness of its mask. The relationship between these parameters is called the aspect ratio. The highest resolution is achieved when the aspect ratio is one. At this ratio, the influence of the mask's sidewalls on the dimensions of the deposited material is minimized, allowing for more precise patterning. In the current work we have used STO-based conducting oxide as an example. The membranes were grown in a Hall-bar shape and then transferred. Transport measurements were performed to study both the electric and magnetic properties.



Optical microscope picture of the transferred Hall-bar shaped freestanding membranes

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Step-edge-induced structure modulation in freestanding BaTiO₃ membranes

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Freestanding oxide membranes offer a promising pathway for integrating devices on silicon and flexible platforms. Freestanding barium titanate (BaTiO₃, BTO) membranes, in particular, hold significant potential for miniaturized field-effect devices integrated on silicon substrates [1,2]. In this study, we investigate the structural modulations in freestanding BTO membranes caused by strain from one-unit-cell step-edges when these membranes adhere to substrates. Using X-ray nanobeam analysis, we examine the (020) Bragg peak with a spatial resolution of 50 nm across varying temperatures. As anticipated, these structural stripes remain rigid above the Curie temperature (120°C for bulk BTO). Our findings highlight that even subtle step-edges, previously overlooked in the study of freestanding membranes, might significantly impact the physical properties of these membranes. Furthermore, the various orientations of the step edges may introduce an additional degree of freedom, potentially leading to the emergence of new properties.

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Can we control the release dynamics of freestanding oxide membranes?

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Over the past few years, sacrificial layer techniques have become a mainstream method for fabricating freestanding oxides [1,2]. One of the drawbacks of this technique is the quality control of the epitaxial growth of oxide heterostructures, which is time-consuming and can usually take several hours. Recently, research has focused on minimizing release times by identifying suitable water-soluble sacrificial layers [3,4]. In this work, we demonstrate the effect of oxygen vacancies on the releasing dynamics of water-soluble $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) layer in the SrTiO_3 (STO) /SAO heterostructures. The STO (~20 nm thick)/SAO (~10 nm thick) films were grown on TiO_2 -terminated STO(001) single crystals by using pulsed laser deposition. To clarify the effect of oxygen-stoichiometry of upper STO films on the underlying SAO layer, various oxygen partial pressure ($P_{\text{O}} = 1 \times 10^{-4}$ to 1×10^{-1} mbar) were employed during the STO film growth while keeping a consistent SAO film growth at $P_{\text{O}} = 5 \times 10^{-4}$ mbar. High-resolution x-ray diffraction (HRXRD) and atomic force microscopy (AFM) were used to examine the crystallinity and surface morphology of the grown films. The water dissolution rate of SAO was determined as a function of P_{O} , revealing a direct correlation between the dissolution time and P_{O} . This study offers valuable insights into how oxygen vacancies influence the release time and structure of SAO, aiding the development of more efficient techniques for fabricating free-standing oxide membranes.

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Thickness controlled spalling for oxide freestanding films via graphene buffer layers

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In recent years oxides materials have attracted considerable attention due to their diverse and remarkable properties, which opens new horizons for sophisticated technological applications. The emergence of freestanding oxides has further revolutionized the scope of these by offering greater flexibility and opportunities to enable the integration of these materials into next-generation flexible electronic devices. Sambri et. al. [Adv. Func. Mater. 30 (2020)] reported the preparation of freestanding $\text{LaAlO}_3/\text{SrTiO}_3$ micro membranes by spalling techniques. However, in spalling the thickness of the resulting freestanding film (FSF) cannot be controlled precisely. Based on recent advances to maintain graphene interlayers during remote epitaxy of oxides via pulsed laser deposition by Ar atmosphere depositions [1], this work investigates in detail the preparation, self-cracking, and structural characterization of $\text{LaAlO}_3/\text{SrTiO}_3$ bilayers on a graphene-coated SrTiO_3 (STO) substrate. Here, the graphene is intended to work as inducing point to terminate the crack along longitudinal direction. The LaAlO_3 (LAO) layer acts as stressor layer on (remote-homoepitaxial) graphene-buffered STO as shown in fig. 1(a), allowing the control the thickness of the resulting FSF.

We successfully prepared the LAO/STO bilayer on graphene coated STO. We also observe that the self-cracking of the bilayer ceases presumably along the graphene layer. Structural characterization by XRD reveals that the LAO/STO bilayer maintain their structural integrity after cracking. However, the LAO film in the graphene-buffered bilayer shows partial relaxation compared to growth of LAO film on single crystal STO substrate as depicted in Fig. 1(b), indicating competing strain relaxation mechanisms [2]. Our findings offer novel perspective for the precise control over thickness during spalling technique.

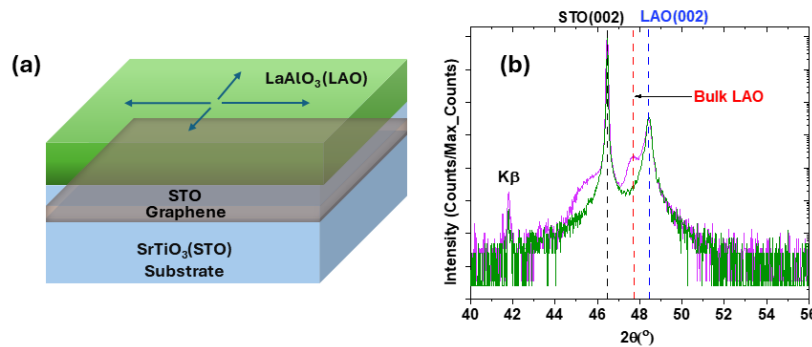


Figure 3 (a). Schematic diagram of heterostructure. LAO is used as stressor layer to apply lateral strain in STO film. (b) Symmetric $2\theta/\omega$ scan of epitaxial LAO film on STO(001)(green) and epitaxial LAO/STO film on graphene/STO(001) (pink) around (002) diffraction peak. Strained (002) diffraction peak and partial relaxed (002) diffraction peak of LAO is marked by blue, and orange dashed line respectively.

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Resonant photoemission spectroscopy investigation of the ferromagnetism enhancement in a $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ membrane free from a SrTiO_3 substrate

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$\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ is a promising half-metallic ferromagnet due to the high Curie temperature (T_C) above 300 K for device applications. Some experimental studies show high performances of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ -based spintronic devices. However, since the magnetization and conductivity of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ are suppressed in the area near interfaces with other perovskite oxides, which is called a “dead layer”, LSMO devices are expected to have a much higher performance originally. Recently, new epitaxial lift-off techniques of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ films from a substrate using a water-soluble layer have reduced the dead layer and improved the magnetization and/or T_C . Clarifying the ferromagnetism enhancement is essential for device applications. We grew a heterostructure of $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (LSMO) / $\text{Sr}_4\text{Al}_2\text{O}_7$ on a SrTiO_3 (STO) (001) substrate by molecular beam epitaxy and transferred the LSMO membrane onto a p-Si substrate [1] [Fig. 1(a)]. Here, we have performed resonant photoemission spectroscopy (RPES) and x-ray absorption spectroscopy to consider the mechanism of the ferromagnetism enhancement in the LSMO membrane. The second-derivative RPES mappings show two Auger components [red dotted lines in Figs. 1(b) and 1(c)]. The enhanced Auger components of the membrane indicate that the strain release from the substrate enhances the itineracy of the Mn-3d electrons, strengthening the double-exchange interaction and improving the ferromagnetism in LSMO [2].

Acknowledgments

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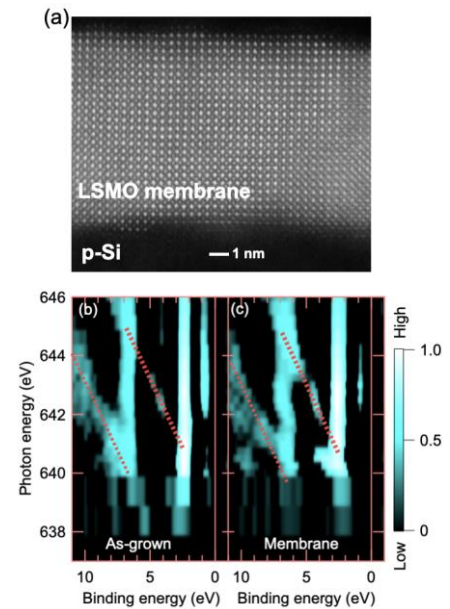


Fig. 1: (a) Scanning transmission electron microscopy image of the membrane. (b) and (c) Second-derivative resonant photoemission spectroscopy (RPES) mappings of the as-grown film and the membrane, respectively. The red dotted lines represent the Auger components.

Bulk micro-machining of SrTiO₃ substrates for device applications

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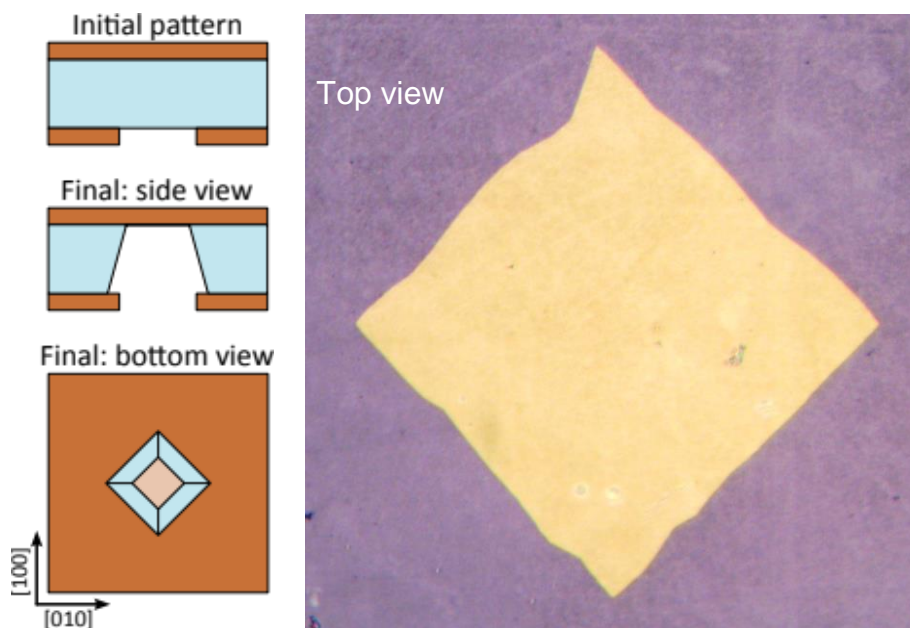
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Advanced fabrication protocols are instrumental to the development of new experiments and device prototypes. For long time complex oxides micro-fabrication was limited to surface micro-machining of thin films by chemical or physical etching or lift-off. More recently, the possibility to exfoliate complex oxide heterostructures from their growth substrate by using a water-soluble buffer layer opened the opportunity to manipulate complex oxides in a similar fashion to Van der Waals materials. Here, we further extend this tool-box by presenting a set of fabrication protocols to realize fully suspended micro-structures from complex oxides thin films by etching through the full thickness of their growth substrate. These methods rely on the selective chemical etching of SrTiO₃ by HF and the use of double-polished STO substrates. These allow for the growth of epitaxial oxide thin films on both sides which are employed as hard masks and/or device layers to realize fully suspended oxide membranes. We will present different examples of possible device geometry, such as trampolines, sealed membranes, or even the substrate itself carved into suspended resonant devices.



(left) Schematic illustration and (right) optical micrograph in reflected light of a 100 nm-thick suspended (La,Sr)MnO₃ membrane obtained by back-side etching of the SrTiO₃(001) substrate

[P2-16]

Phase transitions in freestanding (K,Na)NbO₃ thin films grown by Metal-Organic Vapor Phase Epitaxy

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Potassium Sodium Niobate (K,Na)NbO₃ has gathered significant attention as a promising piezoelectric material due to its excellent piezoelectric and electromechanical properties as well as high Curie temperature (T_c), making it an attractive candidate for high-temperature electromechanical applications [1]. It is well-established that heteroepitaxial film growth on lattice mismatched substrates can modify phase transitions in strained ferroelectric thin films, resulting in properties that could differ from those in the corresponding bulk material. On the other side, chemical lift-off of the functional film from the stiff substrate allows for the investigation of film properties in the unstrained state.

In this study, we report the successful fabrication of freestanding (K,Na)NbO₃ membranes grown by Metal-Organic Vapor Phase Epitaxy (MOVPE) on SrRuO₃ covered DyScO₃ substrates, where SrRuO₃ serves as a sacrificial layer. The membranes were effectively detached by etching the sacrificial layer using sodium periodate. We conducted a thorough investigation of the phase transitions of unstrained (K,Na)NbO₃ membranes, as well as the as-grown (K,Na)NbO₃/SrRuO₃//DyScO₃ heterostack using temperature-dependent X-ray diffraction. Our findings provide insights into the structural behavior of freestanding (K,Na)NbO₃ membranes with significant changes compared to their strained counterpart. Notably, the orthorhombic-tetragonal and tetragonal-cubic phase transitions were detected on the unstrained membranes, which were not observable in the respective strained films.

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[P2-17]

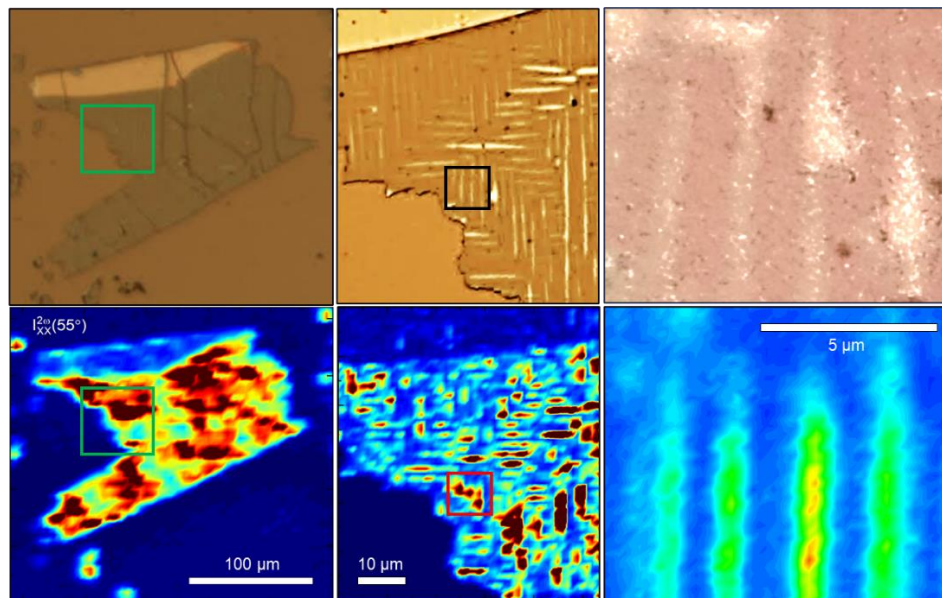
Wrinkle-driven symmetry evolution in $\text{PbTiO}_3/\text{SrRuO}_3$ freestanding membrane

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In this presentation, we report microscopic symmetry study on ferroelectric properties in wrinkled $\text{PbTiO}_3/\text{SrRuO}_3$ (PTO/SRO) freestanding membrane using optical second harmonic generation (SHG) microscopy. A sub micrometer-scale, normally incident beam monitors in-plane symmetry breaking of multi-domain ferroelectric states, i.e., in-plane component superdomain structure, and distinguishes the different combination of superdomain structures. To characterize crystal symmetry of ferroelectric superdomain in the freestanding membrane, we monitor polarization dependent SHG intensity. At the flat surface region, SHG pattern shows two-fold symmetry pattern, which is the result of coherent SHG response of the composed single ferroelectric domain. At the wrinkle structure, on the other hand, SHG responses display two-fold symmetry, but considerably enhances larger 10 times than the flat surface region. In particular, SHG pattern at wrinkle dramatically evolves depending on the direction of wrinkle in a same symmetry. By considering electric-dipole contribution and symmetry analysis, we discuss electrostatic in-plane effect to ferroelectric state in freestanding oxide membrane and its controllability.



Optical microscopy (above) and SHG microscopy (below) images of wrinkled $\text{PbTiO}_3/\text{SrRuO}_3$ freestanding membrane.

[P2-18]

First order polarization process and anisotropic in-plane ferroelectricity in CaTiO₃ thin films

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Antiferroelectric materials are receiving increased attention because of their potential use in high energy density capacitors and electrocaloric devices. These applications rely on their characteristic "double hysteresis" P-E loop, which is caused by a first-order field-induced non-polar-to-polar phase transition [1].

We have discovered a new way of creating double hysteresis behavior in ferroelectric thin films by realizing a first-order polarization process in CaTiO₃, the polar analogue of the first-order magnetization processes (FOMPs) in magnetic materials such as Nd₂Fe₁₄B where higher-order magnetocrystalline anisotropy is relevant [2,3].

While bulk CaTiO₃ is non-polar, tensile epitaxial strain induces a ferroelectric polarization in the plane of the film [4,5]. We can adjust the in-plane anisotropy of the polarization by controlling both strain anisotropy and oxygen octahedral rotation pattern via epitaxial growth on orthorhombic perovskite substrates. By combining electrical measurements, laboratory-based and synchrotron x-ray diffraction, and first- and second-principles simulations, we identify both uniaxial in-plane ferroelectricity and a first-order polarization process in CaTiO₃ thin films.

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Investigating 3D domain structures and superdomains in ferroelectric PbTiO₃ based heterostructures on DyScO₃

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In PbTiO₃ heterostructures epitaxially grown on (110)_o-oriented DyScO₃ substrates with bottom and top SrRuO₃ electrodes [1,2], we previously found that the domain configuration evolves from flux-closure to an a/c-phase as the ferroelectric film thickness increases. Above a critical value of PbTiO₃ thickness, we saw that a modulation in the structure of the top SrRuO₃ electrode appears, demonstrating the possibility of domain nano-engineering in functional orthorhombic perovskites via structural coupling to ferroelastic domains. Moreover, for larger PbTiO₃ thicknesses, we observed a larger scale arrangement of domains into superdomains [1,2].

Based on these observations, we here investigate heterostructures with PbTiO₃ layer thickness ranging from 23 u.c. up to 900 u.c., without the top SrRuO₃ electrode, allowing us to perform direct imaging of the local polarisation by means of piezoresponse force microscopy. We are able to reconstruct the 3D representation of the domain configuration and distinguish among the different superdomains, by combining the information obtained by atomic force microscopy (AFM), vertical and lateral piezoresponse force microscopy (VPFM and LPFM), scanning transmission electron microscopy (STEM) and x-ray diffraction (XRD).

We observe a scaling of the superdomain period with the thickness and note the appearance of superdomain walls, which potentially exhibit switching properties intrinsically different from those of conventional a/c domain walls, and could enhance the functionality of the heterostructures.

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Strain engineering and ferroelectric/antiferroelectric properties of epitaxial perovskite oxide NaNbO_3 thin films

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Antiferroelectric (AFE) materials have stimulated intensive interests owing to their scientific richness and a wide range of potential applications such as electrostatic energy storage capacitors, solid-state cooling devices, thermal switches, displacement transducers and multi-state memory devices [1-2]. Among the AFE class of materials, lead-free NaNbO_3 (NNO) has recently gained attention due to its low-cost component raw materials and rich structural motifs [2]. Moreover, NNO systems also exhibit other superior properties through various external stimuli manipulation. For example, we have achieved giant piezoelectricity in NNO by introducing nanopillar structure in the thin films [3]. This offers opportunities to combine AFE and other properties in developing new multifunctional capabilities in NNO. The AFE phase of NNO, caused by Nb^{5+} atomic displacement and oxygen octahedral rotation patterns, remains stable across a wide temperature range (359°C to -100°C) [2,4]. However, it remains a profound challenge to achieve a stable AFE phase (*Pbcm*, P phase) and a double hysteresis loop is challenging in pure NNO [5]. This is because it tends to favour a more stable orthorhombic ferroelectric (FE) phase (*P2₁ma*, Q phase). Here we comprehensively synthesize and characterize NNO thin films on different perovskite substrates with a large range of lattice mismatch to realize the strain engineering from tensile to compressive. By selecting the substrates with various in-plane lattice constants, the lattice mismatch $\Delta a/a = (a_f - a_s)/a_s$, where a_f and a_s are the in-plane pseudocubic lattice parameters of the NNO films and the substrates, respectively, can be tuned from -2.7% (tensile) to 5.3% (compressive). It is found that the ferroelectric properties are improved in compressive strained NNO films with better polarization-electric field loop. The ferroelectric domain structures could be control through strain engineering. The X-ray diffraction reciprocal space map (XRD-RSM) show that the asymmetric behaviour of NNO thin film structure. The NNO is fully strained at the side of tensile strain, while it is relaxed at the side of compressive strain with lattice mismatch larger than 2.4%. The quarter-integer diffractions are observed on NNO thin films with appropriate compressive strain control, confirming the presence of antipolar structure. More importantly, the size-effect study showed that the double hysteresis loop was observed as the thickness was reduced, further confirming the realization of stable AFE order in NNO thin films. This systematic study on the strain engineering of polarization, domain, antipolar order and crystal structure in NNO thin films will provide important information for further exploration of antiferroelectric materials.

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Zirconium oxide ferroelectrics for memory devices

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Since the first report of robust ferroelectricity in doped HfO₂ thin films in 2011 [1], HfO₂-based thin films have been the candidate material for the next generation of non-volatile memory devices due to the sizeable remanent polarization (P_r) at the nanoscale and good compatibility with silicon-based modern microelectronics, which is of great significance for miniaturization of ferroelectric devices. Chemical modifications play an important role in the property optimization of HfO₂ ferroelectrics. Hf_{0.5}Zr_{0.5}O₂ exhibits robust remanent polarization [2] and some of the best switching performance among HfO₂-based ferroelectric candidates. The new generation of non-volatile random-access memory (NVRAM) devices based on Hf_{0.5}Zr_{0.5}O₂ ferroelectrics are fabricated and attract an increasing enthusiasm from the research community. Given that ZrO₂ is cheaper and more abundant than HfO₂, decreasing the use of HfO₂ will increase the sustainability of this fluorite-structured ferroelectric. In this research, high-quality ZrO₂ nanoscale thin films are prepared epitaxially on differently-oriented SrTiO₃ (STO) substrates by Pulsed Laser Deposition (PLD). The results show that different phases are formed in ZrO₂ thin films depending on the substrate orientation. Both ferroelectric films with orthorhombic phase and non-ferroelectric films with tetragonal phase are observed using XRD and TEM techniques. The thickness limitation to obtain ferroelectric ZrO₂ will be discussed. In addition, ferroelectric-like switching behavior with unexceptional large switching charge is measured in Al/ZrO₂/STO in-plane capacitors. Detailed studies reveal that the polarization loops are not ferroic in origin. Understanding the switching mechanism in ZrO₂ films will help to control the device properties, such as the retention time, of crucial importance for the development of novel memory devices.

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[P2-22]

Controllable periodic domains in BaTiO₃ thin films

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In ferroelectric crystals, spontaneously formed domain structures significantly influence material properties both statically—via changes in macroscopic symmetry and anisotropy—and dynamically, through domain wall motion. Notably, domain size scaling correlates with material behaviors like piezoelectricity and dielectric response.[1] Therefore, engineering the domain type and periodicity could lead to enhanced functionalities. In this work, we demonstrate the high-quality epitaxial growth of BaTiO₃ films on NdScO₃ (110) substrates which provides a low tensile strain state. The flattened energy landscape between different domain variants allows for the emergence of both *a/c*-type domains and unique superdomains. These highly ordered domain structures exhibit satellite diffraction spots around the primary Bragg reflections in the reciprocal space mapping (RSM). Interestingly, domain periodicity isn't solely determined by film thickness; adjustment of growth parameters could result in various periodicities for a certain film thickness, which deviates from Kittel's law and can be potentially related to the spatial period doubling of domains.[2] The switching dynamics and anisotropy of these ordered domains will also be discussed. This work is supported by ARL under W911NF-19-2-0119

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Strain, Young's modulus, and structural transition of EuTiO₃ thin films probed by micro-mechanical methods

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EuTiO₃ (ETO) is a well-known complex oxide mainly investigated for its magnetic properties and its incipient ferro-electricity. Here, we demonstrate the realization of suspended micro-mechanical structures, such as cantilevers and micro-bridges, from 100 nm-thick single-crystal epitaxial ETO films deposited on top of SrTiO₃(100) substrates [1]. By combining profile analysis and resonance frequency measurements of these devices, we obtain the Young's modulus, strain, and strain gradients of the ETO thin films. Moreover, we investigate the ETO anti-ferro-distortive transition by temperature-dependent characterizations, which show a non-monotonic and hysteretic mechanical response. The comparison between experimental and literature data allows us to weight the contribution from thermal expansion and softening to the tuning slope, while a full understanding of the origin of such a wide hysteresis is still missing. We also discuss the influence of oxygen vacancies on the reported mechanical properties by comparing stoichiometric and oxygen-deficient samples.

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Oxygen-driven phenomena in ferroelectric Hf(Zr)O₂ and AlScN thin films – A HAXPES study

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The discovery of a ferroelectric phase of Hf(Zr)O₂ in 2011 and the easily switchable polar state of Sc-doped AlN in 2019 drive the investigation of these ferroelectric (FE) materials, particularly due to their CMOS compatibility and scalability [1,2]. While Sc-doping of AlN weakens the bonds of the polar AlN structure and enables ferroelectric switching below the dielectric breakdown, the “doping” of HfO₂ with oxygen vacancies stabilizes the metastable orthorhombic FE phase. However, the Sc doping of AlN increases its susceptibility to oxidation, while in the more robust Hf(Zr)O₂, the electronic structure is sensitively controlled by filling oxygen vacancies.

The present work explores the electronic structure and chemical properties of Hf(Zr)O₂ and Al_{0.83}Sc_{0.17}N thin ferroelectric films using hard x-ray photoelectron spectroscopy (HAXPES) [3]. For Hf(Zr)O₂, we studied the evolution of the valence band offset with increasing Zr content. Additionally, we assessed the interface stability between various Hf(Zr)O₂ stoichiometries and the TiN bottom electrode [4]. For AlScN, we investigated W-capped and uncapped samples and showed that AlScN is unstable in air due to surface-enhanced oxidation over long periods (weeks to months). By comparing the contribution of Sc and Al relative to the overall oxidation of Al_{1-x}Sc_xN, we assume that oxygen tends to occupy a neighbouring lattice site of Sc, resulting in enhanced oxidation of Sc compared to Al. Moreover, the oxidation process causes N atoms to be removed from their lattice sites, likely leading to an incorporation of interstitial N, deduced from observing a surface-enhanced spectral feature of the N 1s core level [5].

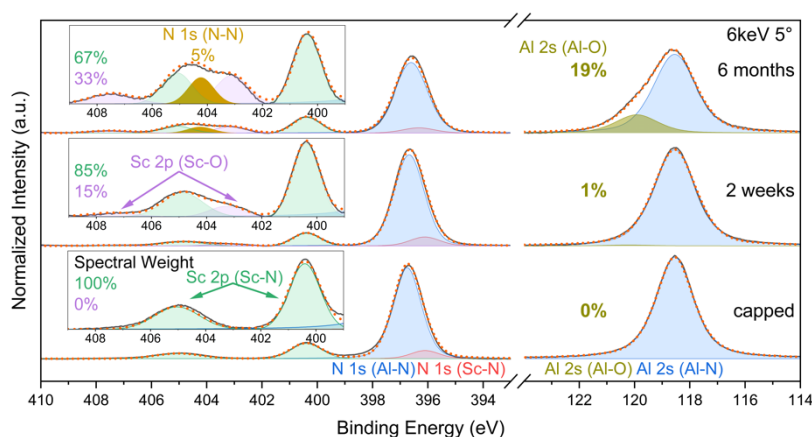


Figure: HAXPES spectra of a 60 nm Al_{0.83}Sc_{0.17}N film, displaying Sc 2p, N 1s, and Al 2s spectra at different stages of air exposure. The spectra were obtained using HAXPES at 6 keV photon energy and a 5° electron emission angle.

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Optimizing HfO₂ based trilayer memristor with ZrO_{2-x} oxygen vacancy reservoirs for advanced neuromorphic computing

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In recent years, the integration of HfO₂ and ZrO₂ materials in neuromorphic computing devices has gained considerable interest within the electronics research community. This study introduces a tri-layer HfO₂/ZrO_{2-x}/HfO₂ memristor, engineered under controlled oxygen partial pressure to establish an oxygen vacancy reservoir in the ZrO_{2-x} layer. This advanced configuration offers remarkable enhanced switching characteristics, exhibiting superior resistive switching behavior, excellent uniformity and durability, and robust multilevel storage capabilities when compared to traditional single-layer and standard tri-layer HfO₂/ZrO₂/HfO₂ memristor. Furthermore, fabricated memristor device has achieved basic synaptic biomimetic functionalities such as, long term potentiation and long-term depression, paired-pulse facilitation and spike-timing dependent plasticity. Conducting the pattern recognition simulation with a neuromorphic network resulted in an impressive accuracy of 92%. We believe that the inclusion of the ZrO_{2-x} layer as an oxygen vacancy reservoir significantly enhanced the electrical properties of the memristor. The results of this research show that the fabricated tri-layer Pt/HfO₂/ZrO_{2-x}/HfO₂/TiN memristor has potential for future applications in non-volatile memory (NVM) and neuromorphic systems.

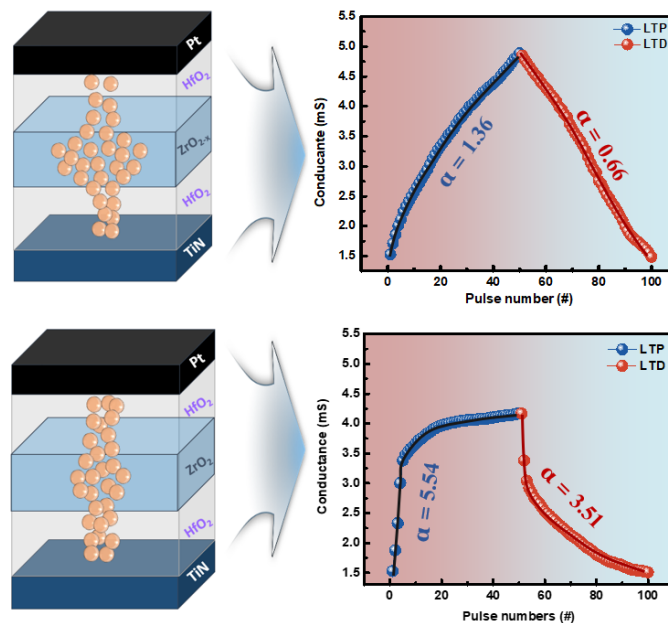


Figure: ZrO_{2-x} oxygen vacancy reservoir layer enhances conductance modulation in HfO₂ based memristor demonstrating improved linearity and performance.

Interface engineering in IGZO-based devicesP. Yuan¹, M. Pereira², J. Deuermeier², A. Kiazadeh², R. Dittmann¹¹ *Peter Grünberg Institut, Forschungszentrum Jülich GmbH, Jülich, 52425, Germany*² *CENIMAT/i3N, NOVA University Lisbon and CEMOP/UNINOVA, 2829-516, Caparica, Portugal*

The reversible alternation of electrical resistance in various metal oxides through electrically induced redox processes is actively investigated for its application in non-volatile data storage and for use in neuromorphic circuits [1]. Typically, conventional memristive devices using a metal-insulator-metal structure show filamentary type switching behavior, necessitating an abrupt electroforming process to create a conductive filament. However, area-dependent type switching devices, utilizing a bilayer structure comprising a conductive oxide layer and an insulating tunneling oxide layer, could avoid local positive feedback caused by inhomogeneous Joule heating during SET process [2]. As a result, this type of devices shows more gradual switching behavior, allowing them to mimic the synaptic behavior in neural networks more effectively [3].

This work aims to fabricate InGaZnO (IGZO)-based area-dependent type switching devices. Initially, the Mo or W bottom electrode layer underwent an oxidation treatment to generate a thin oxide layer [4]. The thickness of this oxide layer is a critical factor influencing the switching behavior, as devices with an insulating oxide layer surpassing the critical thickness necessitate an electroforming process, leading to filamentary type switching. Various combinations of plasma power and duration were examined to yield both low device resistance and a high on/off ratio for area type switching.

Furthermore, both Pt/IGZO and IGZO/MoO_x interfaces form Schottky barriers, with the dominant interface determining the switching polarity of devices. Results indicated that varying the Pt deposition method could modify the upper Pt/IGZO interface, thereby changing the dominant interface and, consequently, the switching polarity. In summary, these IGZO devices could be switched at low voltages while maintaining an on/off ratio above one order of magnitude, accompanied by a narrow distribution of the variability. The full device stack can be fabricated at room temperature without the need for any high temperature growth treatment, ensuring compatibility with CMOS processes and allowing the use of flexible substrates for IoT applications. Additionally, long term potentiation/depression measurements were performed to prove its potential application in neuromorphic computing.

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Area-type resistive switching Pr_{0.7}Ca_{0.3}MnO₃/WO_x devices

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Transition metal oxide (TMO) based memristive devices are regarded as promising candidates for future non-volatile memory devices and artificial synapses for neuromorphic circuits [1]. Although most TMOs exhibit a filamentary type of switching, heterostructures of the quaternary transition metal oxide Pr_xCa_{1-x}MnO₃ (PCMO) and a tunnel oxide layer exhibit a nonfilamentary, area type resistive switching [2]. Due to the lack of a thermal runaway during the SET process, non-filamentary devices exhibit a more gradual switching, that enables the possibility to stabilize intermediate resistance states [3]. This makes the devices highly interesting as hardware implementation of synapses in neural networks. Moreover, the possibility to tune the device current with the area offers new degrees of freedom for the circuit design.

In particular, the reduction of the currents with decreasing area reduces the power consumption for highly scaled devices, which is highly appreciated for neuromorphic computing. As this indicates, in the context of downscaling these PCMO devices it is pertinent to note that while the area dependence is an advantage, it also introduces a caveat when the resistance of the device exceeds a certain threshold, rendering the measurement of the current unfeasible. Since the integration of crossbar structures onto the CMOS chip is the key requirement, the optimization of the device resistance is a critical factor in the downscaling process, while ensuring CMOS compatibility with the fabrication process meaning the constraint that the temperature should not exceed 400°C.

Thus, after a promising material stack was identified with WO_x [4] as the tunnel oxide (TO) different post growth treatment methods of the PCMO depending on the layer thickness as well as growth parameters to reduce the resistance of the devices were investigated further. First, the devices with WO_x were treated with rapid thermal annealing at 400°C or oxygen plasma exhibiting a decrease in resistance. Secondly, the increase of the PCMO deposition temperature to 400°C was investigated resulting in a decrease of the resistance and an increase of the Off/On-ratio. Under these conditions crossbar structures have been successfully established, thus tests of devices integrated on CMOS chips will be presented.

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Electrochemical impedance spectroscopy of artificial protonic synaptic devices

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Biological neural systems effectively process information in real-time using diverse slow dynamics. Replicating these dynamics in scalable electronic devices has been challenging, especially for operations on time scales ranging from seconds to minutes [1]. In this study, we exploited the slow proton dynamics in amorphous WO_3 in 2-terminal protonic device (Fig. 1a) to achieve volatile resistance changes over extended periods. By applying a voltage below 1 V, we induced hydrogenation or dehydrogenation of WO_3 through a Schottky junction with a Pd electrode, resulting in significant changes in electronic conductance (Fig. 1b). While this mechanism successfully facilitated short-term information storage, which is an essential characteristic for implementing online learning with reservoir computing, a deeper understanding of its electric transport behavior was necessary. Especially, it was unclear whether the contribution from the bulk or from the interface is dominant for the proton-induced conduction modulation in Fig. 1b. To address this, we conducted an electrochemical impedance spectroscopy analysis of the device as a function of voltage for the first time. The Nyquist plots (Fig. 1c) displayed two semicircles at low and high-frequency regions, which diminished as a function of DC voltage. Further examination revealed that the first semicircle at high frequency represents the bulk impedance of WO_3 while the second semicircle at low frequency is associated with interfacial impedance. The observed voltage dependence of these two semicircles clearly showed the conductance modulation occurs both in the bulk WO_3 and around the interface simultaneously, leading to the characteristics in Fig. 1b. These findings not only deepen our understanding of the electrical transport in such devices using protons and electrons, but also underscore the potential of this approach for future low-power neuromorphic computing systems, promising substantial advancements in real-time information processing and storage technologies.

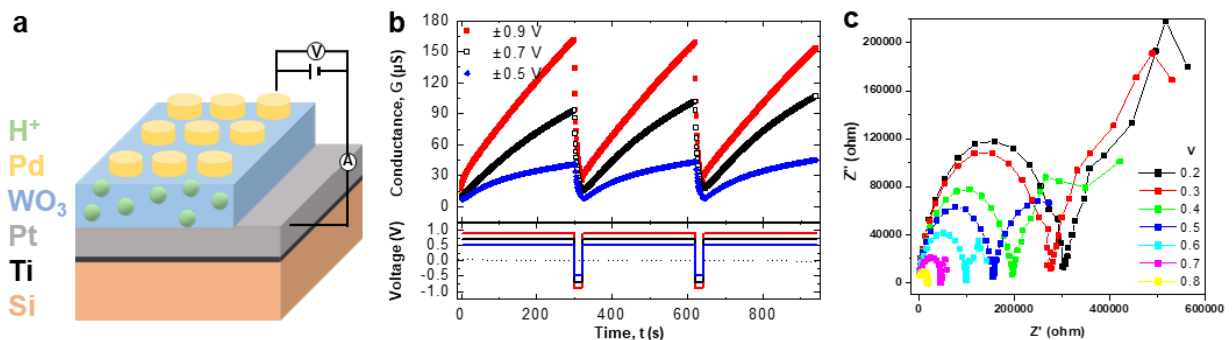


Fig. 1: Schematic representation of (a) 2-terminal protonic device. (b) Switching of protonic conductance by electrode voltage. (c) Nyquist plots of (a) as a function of DC voltage after exposure to H_2 . The frequency range was from 100 kHz to 100 mHz.

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Review of the proposed switching mechanisms in $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ heterostructures considering the recent process with amorphous PCMO

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Heterostructures of the perovskite $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ (PCMO) and a tunnel oxide are a well-studied system and a famous representant of area-dependent switching in the family of valence change memory [1]. As the valence change by oxygen migration is in this system mainly field driven and not as temperature driven as for filamentary devices, both switching directions set and reset can be tuned gradually, which makes them highly interesting for the application in neuromorphic circuits [2], [3]. Further PCMO based devices are of serious interest for the memory industry in DRAM like persistent memory. Recently the publicly listed company 4DS memory claimed to achieve with PCMO based devices sub 5 ns writing speed with an endurance of over 10^9 cycles. Explaining the switching mechanism different effects are considered by the movement of oxygen, considering the changing of conduction over the tunnel oxide and the change in conduction over the area near the interface in the PCMO. A decrease of conduction in the interface region in PCMO is explained by a reduction of the carrier mobility by the introduction of oxygen vacancies [4], an increase of the band gap [5] and by the change of a space charge region by the change of charge carrier density by oxygen vacancies [2], [6].

The above discussed switching mechanisms in the PCMO are proposed for devices with crystalline PCMO layers. The urge to use CMOS BEOL compatible temperatures with limits of 400°C for the PCMO deposition resulted in progress of resistive switching with amorphous PCMO layers since 400°C is below the crystallization temperature. The finding of good switching amorphous PCMO layers raises questions for the validity of the proposed switching effects in the PCMO since the previously made assumptions on the electronic structure are challenged. The switching effect can neither be reduced to only considerations based on variation of the transport over the tunnel oxide since the switching differs strongly between amorphous PCMO layers with different deposition temperatures. Therefore, reviewing the switching mechanisms under consideration of the new findings for amorphous PCMO gives a valuable new perspective on the industrial, highly interesting class of PCMO based memristive devices.

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Remotely triggered resistive switching in multiterminal VO₂ devices

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In resistive switching, a device is switched between two or more resistance states by (pulsed) external stimuli, typically of electric, optical, or thermal origin. Devices with this property are of interest for rapidly developing, energy-efficient computing paradigms – e.g., in-memory and neuromorphic computing – envisioned to meet the computing demands of the AI revolution in a sustainable manner.

A variety of (often intertwining) material properties and physical processes underlies resistive switching behavior: for example, ionic transport, redox reactions, ferroic domain order and motion, spin alignment, structural phase transitions, electronic phase transitions, and filament formation [1]. Developing and applying an in-depth understanding of these properties and processes allows engineering new functionality in devices with specially designed geometries.

Here, we expand the typical two-terminal device geometry by adding a third electric terminal, which enables triggering remotely a resistive switch between the two other terminals. This effect hinges on the thermal hysteresis, filament formation, and reversibility inherent to the electrically triggered insulator-to-metal transition in the active material, VO₂ [2]. Contrary to local resistive switching between any two terminals in our device, the remotely triggered switch persists after the stimulus on the third terminal is removed. Both current and voltage as well as mixed stimuli can trigger the remote switch, allowing same-quantity gain or voltage-current interconversion. The remote effect is observed if a ‘drain-source’ bias is applied within the hysteresis loop observed for two-terminal switching. In the middle of this region, the change in channel resistance induced by the remote switch is maximized at ~2500%.

Multiphysics simulations show that the thermal crosstalk between filaments – previously observed for parallel two-terminal devices [3] – drives the remote switch. Moreover, tuning the applied biases enables controlling the pathways taken by the filaments, realizing filament patterns that depend on the device history in a complex manner. These devices pave the way for realizing reconfigurable logic [4] inherently at room temperature and provide a multidimensional platform for studying the dynamics of the electronically triggered metal-to-insulator transition in VO₂ and related materials.

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High temperature cluster spin glass behavior in defect controlled $\text{Y}_3\text{Fe}_5\text{O}_{12}$ thin films towards neuromorphic computation

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A spin glass (SG) is characterized by a frozen spin state at low temperatures, resulting from competing randomness and frustration of magnetic interactions. This state also realized photoinduced magnetism and an aging memory effect: that creates a history-dependent property. This unique feature presents an opportunity to replicate the functions of the human brain, oblivescence a similar Hamiltonian framework. We aim to study the spin dynamics based ultra-low power data carrier (magnon) in the spin glass system at frozen state to implement a reservoir computing system mimicking the ultra-low power operation of the human brain. However, the spin frozen state in spin glasses (SG) only manifests at extremely low temperatures, limiting their practical and low-power applications. Consequently, this research primarily aims to explore the potential for developing optimal room temperature spin glass materials by manipulating the disorder within garnet-based ferromagnetic materials. Considering magnetization dynamics for further study, we began with yttrium iron garnet (YIG: $\text{Y}_3\text{Fe}_5\text{O}_{12}$) due to its exceptionally low Gilbert damping constant.

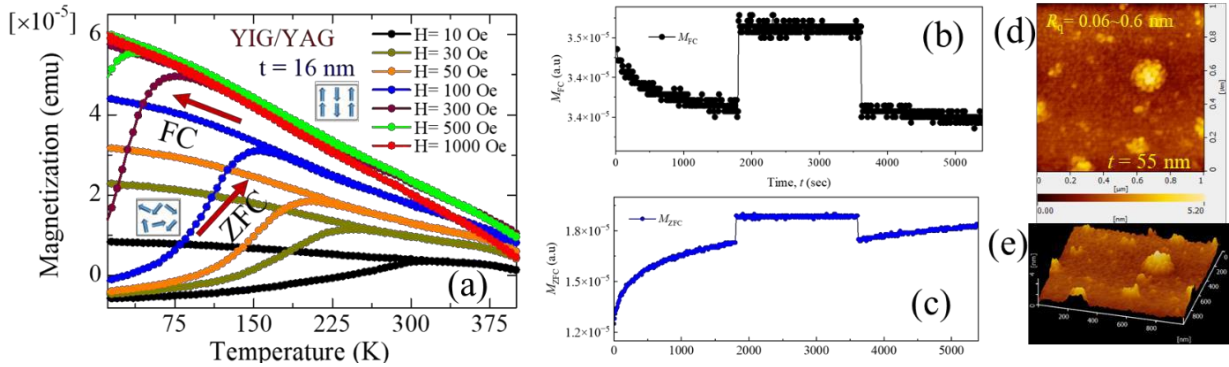


Figure 1. (a) Zero field cooling (ZFC)-Field cooling (FC) behavior under different DC magnetic fields in YIG/YAG films. Characteristics of aging memory effect for (b) FC and (c) ZFC. (d) 2D and (e) 3D AFM images of the film surface signifying the surface modification due to the crystal dislocation effect.

We have prepared 16 nm thick YIG thin films on the (001)-oriented YAG substrate using pulsed laser deposition. We have investigated field cooling (FC) and zero field cooling (ZFC) properties of the YIG/YAG using a standard protocol and have observed the characteristics of bifurcation behavior at near room temperature (see Fig.1(a)) compared to a previous report [1]. We have calculated the Mydosh parameter as $k = \Delta T_p / T_p \Delta(\log_{10} f) \sim 0.023$ signifying the material as cluster spin glass. We further have confirmed the characteristic aging memory effect for negative heat cycle using standard FC-ZFC protocol which is shown in Figures 1(b) and (c). Magnetization relaxation can be used to fade memory for reservoir computation. We have identified the dislocation induced defect as the origin of the presence of frustrated spin state in the ferromagnetic YIG film. As the critical thickness of YIG/YAG is less than 10 nm due to a large compressive lattice mismatch (2.9%), the film relaxes during growth. We have confirmed this by RSM mapping. This film relaxation generates a significant amount of dislocation, one of whose pieces of evidence is extremely rough surface morphology shown in the atomic force microscopy imaging in Figure 1(d) and (e). These dislocations result in frustrated states which induce the spin freezing effect at room temperature. Further investigation will be conducted using XMCD and STEM measurements to confirm the crystal deformation regarding orbital momentum and atomic imaging, respectfully.

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Non-volatile electro-thermal memristive behaviour in planar NdNiO₃ thin films

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Memristive two-terminal devices are the subject of intensive research in the emerging field of neuromorphic computing. They are investigated as hardware primitives for artificial neural networks that emulate essential functionalities of their biological counterparts. One of those functionalities is the capability to store synaptic weights via non-volatile resistive switching.

Here, we report on non-volatile, multi-state electrothermal memristive switching in charge-transfer insulator NdNiO₃ (7 nm thick) thin films. We combined electrical transport measurements with optical wide-field microscopy of the metal-insulator phase separation. We found resistive switching via multiple persistent states accompanied by the formation of a metallic filament between the two device electrodes. We interpret these results with electrothermal instability and conclude that this unexpected behaviour results from a distinct hysteresis in the resistance vs. temperature dependence of the NdNiO₃-thin films.

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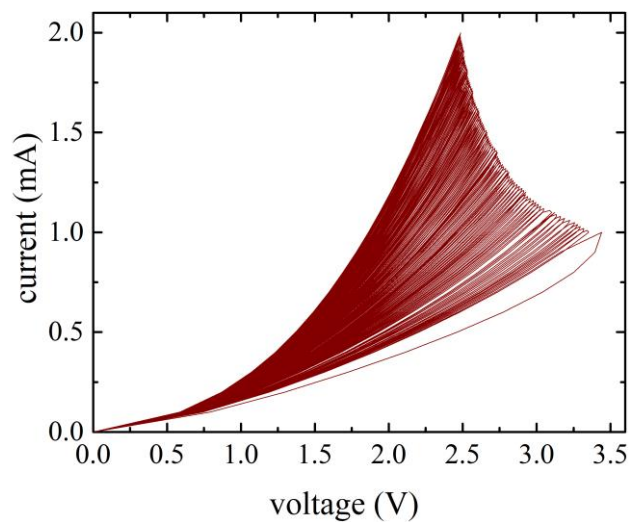


Figure 4: Series of current-voltage loops demonstrates the information storage capability of the NdNiO₃ thin film. Maximum current increased by 0.01mA steps after a loop is completed.

[P2-33]

All HfO_x resistive switches with artificial virtual electrode and self-limited oxide layer

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Resistive random-access memory based on hafnia is a potential candidate for next generation memories. However, there are some key challenges to overcome including the stochastic nature of the filament formation during electroforming and SET operation which results in large cycle-to-cycle and device-to-device variability. To reduce the stochasticity of filament growth, we propose a simplified device concept with an artificial virtual electrode layer of oxygen-deficient HfO_x with a well-defined self-limited oxidized switching layer. Oxygen-deficient virtual electrode device stacks (TiN/HfO_x/HfO₂/Pt) were grown by adjusting the correct oxygen content in the virtual electrode through precise control of the oxygen flow during MBE growth, resulting in an oxidation of these layers in an oxygen rich atmosphere. The depletion of oxygen turns the electrodes into an oxygen-vacancy exchange layer, which stabilizes the filament in the thin switching layer. Detailed electrical characterization demonstrates that the artificial virtual electrode layer combined with the self-limited oxidizing switching layer can significantly reduce electroforming and SET voltage stochasticity and thus improve the switching reliability.

Growth conditions and characterization of SrCoO_{3-δ} thin film protonic conductor for electrochemical resistive switching

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There are currently essential scientific efforts to bridge artificial intelligence (AI) and the human brain. Memristive devices, which change electronic conductance by electric modulation, share remarkable similarities to biological synapses, dendrites, and neurons at both the physical mechanism level and unit functionality level, making the memristive approach to neuromorphic computing a promising technology for future sustainable artificial intelligence. Electrochemical resistive switch (ERS) is most suitable for symbiosis of artificial intelligence (AI) and human brain as the connection between a neuron and a human cell is provided via chemical signal and/or electrical signal transfer carried by ions between cells.

Our conceptual approach is the development of the novel proton-based ERS devices compatible with complementary metal-oxide-semiconductor (CMOS) technology. Importantly, proton exchange is natural for human cells and request lower energy consumption and higher operation speed due to the small H⁺ radius.

Here, we demonstrate our first achievement on the development of a perspective protonic conductor SrCoO_{3-δ} (SCO) [1]. SCO thin films are grown by using pulsed laser deposition at various deposition parameters such as oxygen pressure, etc. The chemical composition, electronic structure and oxidation state of elements of as-prepared SCO, as a function of growing conditions, as well as chemical compatibility between SCO thin films and different substrates, are *in-situ* studied by X-ray photoelectron spectroscopy (XPS) in the DAISY-BAT cluster tool [2]. The impact of the oxygen content on the crystal structure variation of SCO thin films is studied by X-ray diffraction (XRD). Morphology and thickness of the SCO thin films are analyzed by using scanning electron microscopy (SEM) and X-ray reflectivity (XRR).

Our results evidence the direct correlation between the oxidation state of Co ions, stoichiometry and crystal structure of SCO thin films. The approaches to design the SCO films with a desirable oxygen content are discussed.

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Stable conductance modulation in WO₃-based proton devices via O₂ removal

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The control of physical properties using hydrogen in oxides has attracted much attention, and it is expected that functional devices will be fabricated by combining such materials in the heterostructures. Among various oxide materials, tungsten oxide (WO₃) has long been known as a mixed conductor of electrons and protons and is regarded as an important component of hydrogen-based functional devices. Previous studies have shown that hydrogen can be volatily modulated in and out of the WO₃ thin film from a hydrogen-absorbing Pd electrode, leading to volatile conductance modulation which can be used as short-term memory in neuromorphic functions. However, hydrogen-based devices are intrinsically unstable, and the conductance changes over time. In this study, a systematic evaluation of the influence of the measurement atmosphere revealed that oxygen in the atmosphere is the main cause of instability, and that stable electrical characteristics can be obtained over a long period of time if oxygen is eliminated in a vacuum.

In the experiment, a two-terminal device with a Pt layer of 30 nm, an amorphous WO₃ layer of 120 nm, and a Pd layer of 30 nm was fabricated on a silicon substrate (Fig. 1a). When hydrogen is absorbed in Pd in a hydrogen atmosphere and then a positive pulse voltage is applied to the Pd electrode after removing it from the hydrogen atmosphere, the WO₃ is hydrogenated by electrochemical reaction and the electrical conductance of the device increases. This change in electrical conductance is volatile and is reset to the initial state when the voltage is dropped to near 0V. The problem is that the electrical conductance is not stable in air. For example, as shown in Fig. 1b, when left in air, the electrical conductance gradually decreases due to the loss of hydrogen. Furthermore, the wake-up effect is also observed, in which the electrical conductivity gradually increases after repeated measurements. On the other hand, in vacuum, the electrical conductance is stable, and changes only a little even when the sample is left for several days or when the pulse voltage is applied repeatedly. Furthermore, measurements in nitrogen and in dry air showed that the instability of the electrical conductance was caused by oxygen, and that the stable properties could be maintained by removing the oxygen from the atmosphere.

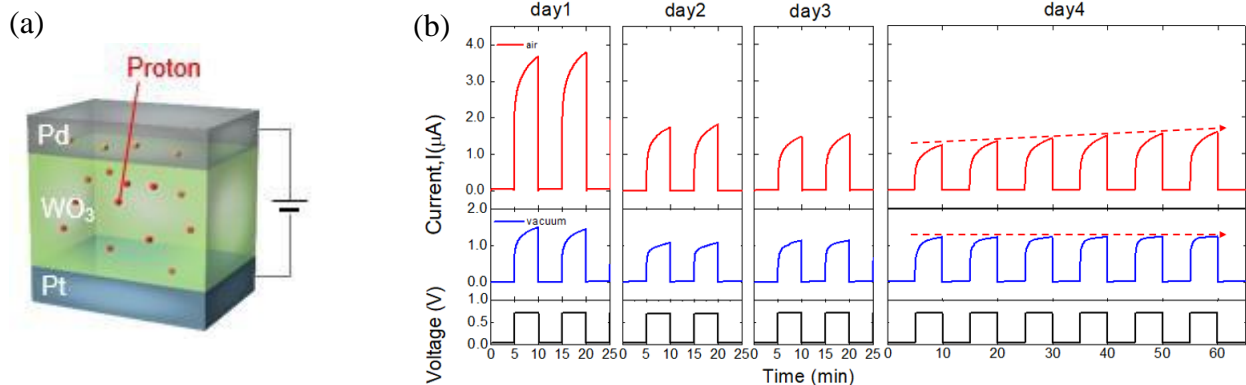


Fig.1(a) A two-terminal device using protons. (b) Current modulation by pulse voltage in the air and in the vacuum

Influence of hafnium oxide layer thickness on the performance of Cu/HfO₂/Pt CBRAM device

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This study investigates the impact of hafnium oxide (HfO₂) layer thickness on the performance of conducting bridge random access memory (CBRAM) devices tailored for two terminal selector devices. Samples with varying HfO₂ layer thicknesses (20 nm, 15 nm, 10 nm, 7 nm, 5 nm, and 3 nm) utilizing a Pt bottom electrode and a top electrode stack (Cu/Pt/Au) were fabricated and thoroughly characterized.

Utilizing molecular beam epitaxy (MBE) for HfO₂ layer deposition ensured superior crystallinity and minimized energy grain boundaries, crucial for facilitating Cu ion diffusion paths. Samples were categorized based on thickness, revealing distinct electrical characteristics. Thinner samples (10 nm, 7 nm, 5 nm, and 3 nm) exhibited lower set voltage (~1 V) and resistance at the high resistance state (HRS), whereas thicker samples (20 nm, 15 nm) demonstrated higher and broader set voltages (~4 V) and elevated HRS resistance.

Further analysis of the reset process revealed variations in maximum current flow, with thicker samples exhibiting higher currents (3 mA) compared to thinner counterparts (1 mA), suggesting potential concerns regarding joule heating. Quantum point contact (QPC) modeling provided insights into energy barrier heights and widths between ruptured conducting filaments. Specifically, 20 nm and 15 nm samples exhibited high energy barriers (~2.5 eV), while the 10 nm sample displayed a lower energy barrier height (~1 eV) and smaller barrier width (0.7 nm), indicating distinct conduction mechanisms.

These findings underscore the critical role of HfO₂ layer thickness in modulating resistive switching behavior and electrical conduction mechanisms in Cu/HfO₂/Pt devices. The proposed models enhance our understanding of conducting filament behavior, offering valuable insights for optimized device design for two terminal selector devices.

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Electric-field control of the local thermal conductivity in negative charge transfer oxides

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The dynamic control of the thermal conductivity of crystalline solids remains one of the biggest challenges of materials science. So far, the most successful strategies exploited the electric/magnetic field control of ferroic polarization in ferroelectrics and magnetic materials [1,2], or topochemical ion exchange in transition-metal oxides [3,4]. However, the small thermal contrast, poor cyclability or the lack of spatial control over the effect, remain serious issues to be solved. Here, we report the continuous tuning of the local thermal conductivity in Mn, Fe and Co oxoperovskite thin films by a voltage-biased AFM tip, at room temperature. Micron-size domains with a defined value of the thermal conductivity can be “written” by this method, achieving reductions as large as 50%. The thermal states are stable under normal atmospheric conditions, but can be returned to the original state by thermal annealing in air. The method presented here constitutes a sort of AFM lithography of thermal states and opens the door to the fabrication of tunable elements of a thermal circuit.

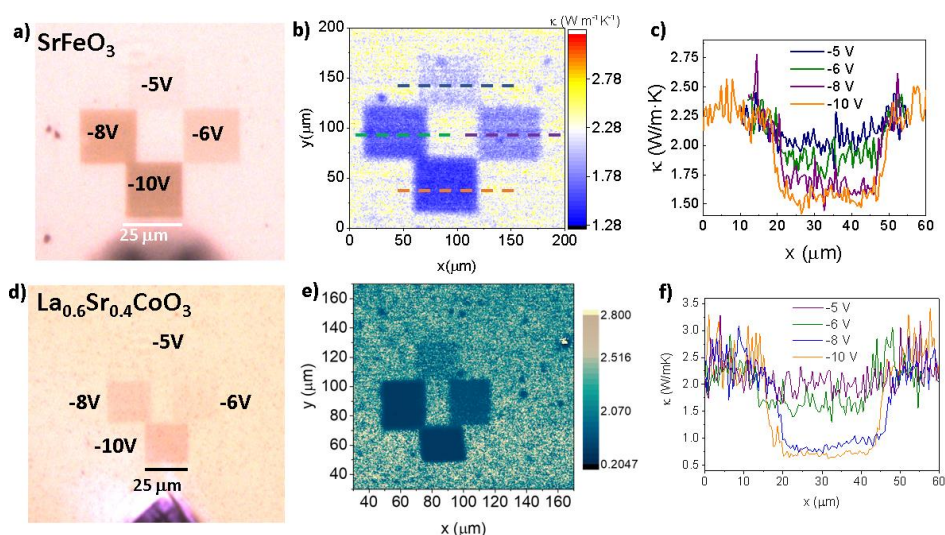


Figure 1. a): Optical image of the surface of a SrFeO_3 thin film, with pads of 25×25 microns previously scanned with a voltage-biased AFM tip. b) Thermal conductivity map obtained from Frequency Domain Thermoreflectance experiments. c) Thermal conductivity line scans along the pads in b). Similar experiments are shown in d-f) for $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_3$.

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[P2-38]

Nano engineered solid state ionic metal oxides for near-room temperature oxygen conductivity

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Solid State Ionics (SSI) is the study of devices and materials with ionic, electronic mixed and fully ionic conductors' properties. Oxide ion conductors (OCs) are an important class of materials with key technological applications for sensors and electrochemical energy systems. The present work is interested in exploring fast oxygen ions transport at their interface and electrochemical reactions in near-room temperature domain in nanoscale (thin films around 100 nm) for the 2D-layered heterostructure hexagonal Perovskite $\text{Ba}_7\text{Nb}_4\text{MoO}_{20}$ (Bulk) due to its structural features with the low activation energies (0.56 eV) through an interstitial oxygen migration leads to the conductivity in the range of $5 \times 10^{-4} \text{ S cm}^{-1}$ at 350°C [1,2]. The epitaxial growth of thin film is achieved by pulsed laser deposition (PLD) technique and in-situ electrochemical impedance spectroscopy (EIS) technique would be the major characterization to understand and improve the oxygen transport at the interface side of this complex structure in the nanoscale range to design it as a solid-state electrolyte for metal-air batteries application.

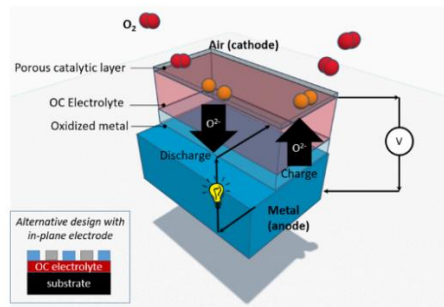


Figure 1: Oxide ion conducting Solid-state electrolyte for Metal-air batteries.

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Resonant $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ suspended microbridges: mechanical characterisation, integration of the piezoelectric layer and application as bolometers

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In resonant bolometers, the power of the incident radiation is measured via a frequency shift of the mechanical resonance of a suspended structure. In this work, we propose to integrate a $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$ (PZT) piezoelectric layer both for actuation and reading, on top of epitaxial LSMO thin films deposited on buffered silicon substrates. First, pulsed laser deposition was used to grow the 45 nm thick $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (LSMO) thin films on CaTiO_3 (CTO) layers of different thicknesses (in the 5-42 nm range) on top of 20 nm thick SrTiO_3 (STO)-buffered silicon substrates previously prepared by molecular beam epitaxy [1]. As shown in Fig. 1(a), on each $5 \times 10 \text{ mm}^2$ Si substrate, 66 groups of 9 suspended bridges of width 6 μm and of length ranging from 25 μm to 250 μm were fabricated using a combination of ion beam etching (IBE) and reactive ion etching (RIE) with SF_6 [2]. Second, the samples were glued on a commercial piezoelectric plate and the resonant frequency of each resonator (suspended microbridge) have been measured in the 400-1300 kHz range by an optical level system [3]. We extracted the distribution of the stress by using the string resonator model [4]. A very high Q factor was measured by amplitude decay in the 20000-120000 range. It was demonstrated that the CTO layer thickness can be successfully used to control the strain in the films. For example, Fig. 1(b) shows the very uniform stress map in the LSMO/CTO/STO/Si suspended bridges that we could obtained with a 14 nm thick CTO layer. The variation of the resonant frequency with temperature were also investigated, which gives an estimated value of the sensitivity of the final devices [5]. Finally, a 150 nm layer of PZT was deposited by sputtering at room temperature followed by rapid thermal annealing on selected LSMO/CTO/STO/Si samples. Subsequently, IBE and RIE steps complete the full process of the final device, integrating the PZT layer. Finally, this piezoelectric self-actuation was tested, including the PZT electrical and mechanical properties.

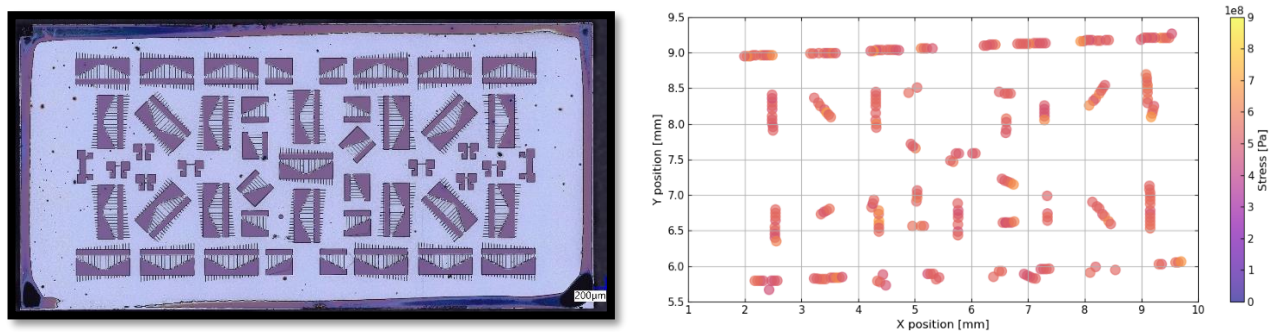


Fig. 1 : (a) Optical microscope photograph of a $5 \times 10 \text{ mm}^2$ Si substrate containing suspended bridges of various lengths; (b) Corresponding stress map of a LSMO/CTO/STO/Si sample with 14 nm thick CTO.

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Electrically tunable free space barium strontium titanate (BST) optical filters

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Barium Strontium Titanate ($\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$) with different ratios of Ba/Sr [1] has been widely used in the microwave regime where permittivity tunability is one of the essentials in Varactors [2]. Here we present a BST-based optical filter. A 350 nm thick BST film sandwiched between two highly reflective distributed Bragg mirrors (DBR) forms a Fabry- Pérot resonator with an electrically tunable cavity. The filter offers a stop band with a width of 400 nm while it becomes transmittive at the resonance frequency. The initial result in Figure 1.a shows the transmission notch measured with a reflectometer.

Applying an electrical field through planar electrodes will alter the optical length of the cavity and thus the notch wavelength. Figure 1.b indicates the simulation result with only $\pm 1\%$ optical path length difference. Experimental confirmation is in progress.

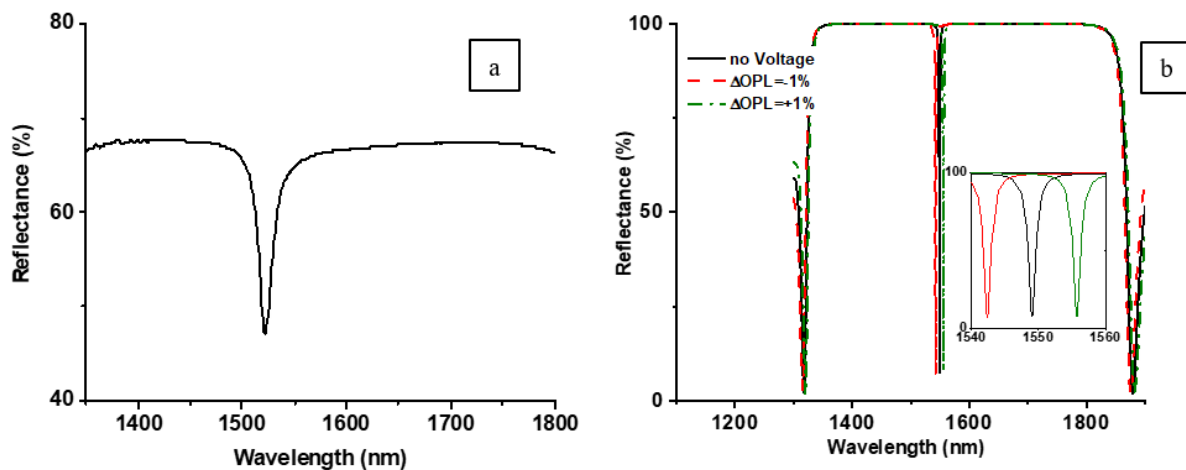


Figure 1. a) Notch filter at 1522 nm. b) Tunability of notch wavelength with the change of optical path length (OPL)-inset is a closer look at the notch wavelength

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Freestanding SrMoO₃ membrane single layers and complete devices: strain mechanisms and high-performance varactors

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Freestanding membranes fabricated by dissolving sacrificial layers have emerged as a cutting-edge approach in materials science, particularly for complex oxide thin films. Traditional epitaxy, which involves growing films on substrates with similar lattice parameters, imposes significant constraints on material properties due to lattice mismatch and substrate clamping effects. This research aims to overcome these limitations, fundamentally studying the strain mechanisms in epitaxial films and exploring the possibility of transferring functional oxide devices to Si wafers.

The highly conductive perovskite SrMoO₃ exhibits remarkable low room-temperature resistivity, down to 5.1 $\mu\Omega\cdot\text{cm}$ [1] in single crystals and below 20 $\mu\Omega\cdot\text{cm}$ [2,3] in thin films, making it an ideal bottom electrode for functional oxide thin films. Using the freestanding thin-film lift-off technique, we investigated the strain mechanisms in SrMoO₃ layers grown epitaxially on scandate substrates, achieving thicknesses beyond the conventionally estimated critical thickness, up to several micrometers [4]. Comparative analysis of lattice constants revealed that SrMoO₃ is elastically strained within the heterostructure and fully relaxes after substrate removal. A minor trend of strain-induced off-stoichiometry was observed, leading to an expansion of the actual bulk lattice parameters of freestanding SrMoO₃ films [5].

Going beyond single layers, we successfully transferred a complete device, namely a freestanding all-oxide varactors based on (Ba,Sr)TiO₃, using SrMoO₃ as a bottom electrode, to Si substrates. These varactors exhibit low losses at high frequencies (1–3 GHz) and can be effectively tuned at a Li-ion battery voltage of 3.7 V. A leakage current density as low as 1 A/m² is achieved for dielectric layers below 50 nm with 1 at. % Mn doping. This makes these varactors highly relevant for high-performance agile devices, offering low tuning voltages and ultra-low power consumption.

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All-oxide thin-film varactors with SrMoO₃ bottom electrodes and Mn/Ni-doped BST for sub-6 GHz applications

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Perovskite materials are ideal for applications in oxide electronics due to their rich phenomenology of functional properties. The advantage of perovskite type oxide bottom electrodes in electronic devices lies in the possibility to grow the successive isostructural functional perovskite layers epitaxially with low defect densities using conventional thin film deposition techniques. Realization of all-oxide electronic devices requires highly conducting oxide electrode materials, which can be integrated into the device structure. In the previously investigated all-oxide metal-insulator-metal (MIM) ferroelectric tunable capacitors (varactors), an epitaxial functional oxide dielectric Ba_xSr_{1-x}TiO₃ (BST) layer is grown over a micrometer-thick highly conducting optically transparent electrode thin film of SrMoO₃ with low room-temperature resistivity of only 20-30 μΩcm [1, 2, 3]. The high electric conductivity of SrMoO₃ originates from the $4d^2$ electronic configuration of Mo⁴⁺, resulting in a large charge-carrier density beyond 10²² cm⁻³ and suppressed scattering due to the energetic separation of the *s*- and *d*-states.

In the MIM all-oxide varactor geometry, a permittivity of the BST layer is tuned by an electric field, applied between the bottom SrMoO₃ and top Pt electrodes. A small thickness below 50 nm of the BST functional layers is accessible due to low defect density, enabling high tunability of 60% at low tuning voltages of the varactors at the level typical for Li-ion batteries of 3.8 V [3]. However, a low Schottky barrier at the interface between two isostructural perovskite oxides SrMoO₃ and BST results in high leakage currents in the varactors, causing significant losses that negatively affect the quality factor and overall performance of the varactors [3]. The leakage currents originate from a high concentration of oxygen vacancies in the BST layers which have to be grown at non-optimal low oxygen pressure to prevent oxidation of the underneath SrMoO₃ electrode films to an insulating SrMoO₄ with Mo⁶⁺ [4].

Here we present the results of our recent studies of all-oxide varactors with the BST functional layers doped with Mn and Ni [5]. The performed XANES investigation of the doped BST films revealed the Mn²⁺, Mn³⁺ and Ni²⁺ dopants with small ionic radii, which occupy Ti⁴⁺ sites of similar size in the BST perovskite structure and act as acceptors, neutralizing oxygen vacancies. We observe a drastic suppression of the leakage current at room temperature by more than 5 orders of magnitude down to 1 A/m² at 100V/μm with only 1% Mn doping. This result indicates nearly full {Mn²⁺V_O} association of oxygen vacancies which are naturally present in the BST films of the Pt/BST/SrMoO₃ all-oxide varactors, grown at low oxygen pressure.

The doping approach to reduce the leakage current enables realization of the all-oxide varactors with the functional doped BST layers thinner than 30 nm. An exemplary all-oxide varactor with a 25 nm thick BST layer exhibits a quality factor of up to 40 at 1 GHz and a tunability of 55% at a low voltage of only 2.9 V. Beyond the suppression of the leakage current, BST doping was also found to change the acoustic properties of the material, damping the excitation of the acoustic resonances and shifting the resonance peaks to higher frequencies.

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(La_{0.7}Sr_{0.3})MnO₃ trampoline resonators for magnetometers and IR bolometers

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We present suspended devices made from (La,Sr)MnO₃ (LSMO) single-crystal thin films deposited on top of SrTiO₃ single-crystal substrates by Pulsed Laser Deposition. We show different examples of micro-mechanical resonators, such as cantilevers, double-clamped bridges, or trampolines realized with specific fabrication protocols and having different shapes and characteristic lengths up to 1 mm. At first, we perform magnetic characterization and near-IR optical spectroscopic ellipsometry measurements. We then analyze the mechanical properties of the LSMO suspended structures in terms of stress, mechanical quality factor (Q), and responsivity to incident light. The specific magnetic and optical properties of LSMO make these devices promising candidates to realize high-Q resonant magnetic sensors or mechanical infrared bolometers working at room temperature.

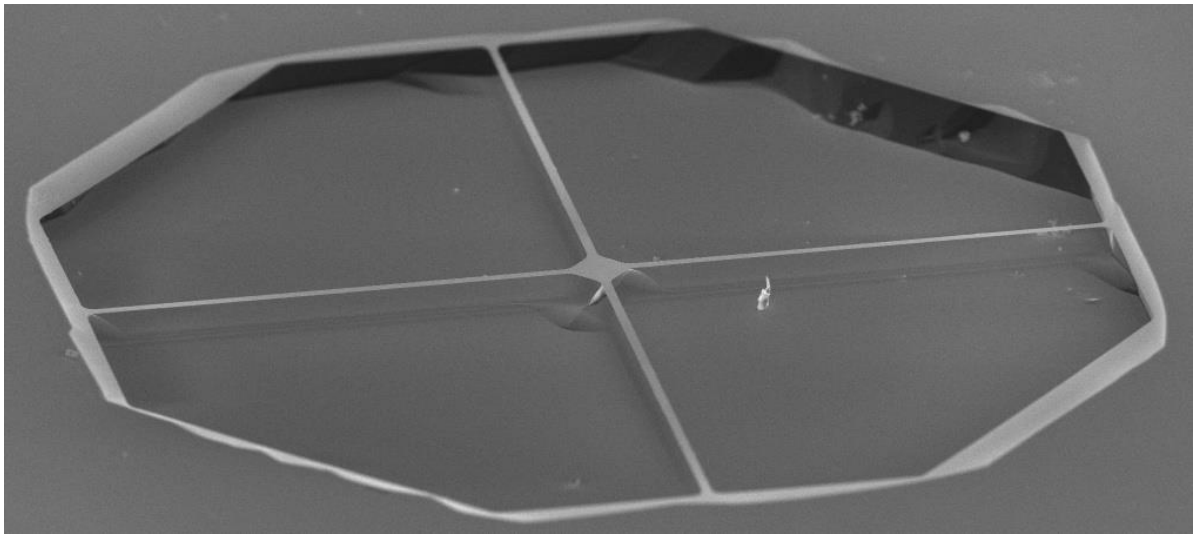


Figure 1: (La,Sr)MnO₃ suspended trampoline resonator.

Nanomechanical probing of the elasto-plastic responses of thin film SrTiO₃

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Oxide thin films play a crucial role in oxide electronics. Defects of various dimensions in oxide films, particularly point defects such as oxygen vacancies and line defects as dislocations, can directly impact the functional stability and mechanical reliability of the electronic devices. Such defects can be generated unintentionally during the fabrication or the packaging process. Developing effective assessment approaches for probing such defects generation is useful to unleash the technological potential of functional oxide thin films. Here, we propose a nanomechanical probing approach to assess the elastic to plastic deformation responses of thin film SrTiO₃ deposited on SrTiO₃ substrate, which serves as a model material. To verify the sensitivity of this nanomechanical probing method, we fabricated the thin films with three different non-stoichiometries and defects densities in 1) vacuum, 2) oxygen-rich environment, and 3) a mixture of both, using pulsed laser deposition. Upon shallow nanoindentation tests, the three thin films exhibit different incipient elastic to plastic transition behavior. Using X-ray diffraction analysis (XRD), X-ray photoelectron spectroscopy (XPS), and transmission electron microscopy (TEM) analysis, we correlate the defects type (point defects and dislocations) with the nanomechanical responses. Our approach of using nanomechanical testing may serve as a new route for fast probing of defects in thin films.

Keywords: thin film; dislocations; nanoindentation; non-stoichiometry

Epitaxial NiFe₂O₄ films model systems for studying the inverse spinel facet OER activity

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Nowadays, with the growing focus on green energy sources such as solar cells and wind power, the ability to store the excess energy for later use has become essential for utilizing it as efficient as possible. One of the most promising approaches is storing energy in the form of chemical bonds – hydrogen. One way of producing it via non-CO₂ emitting technologies is water electrolysis, however one of the half-reactions – oxygen evolution reaction (OER) suffers from sluggish kinetics and remains a bottleneck of water electrolysis [1]. Spinel oxides are a promising class of materials that is being actively studied for OER catalysis application due to consisting of earth-abundant, inexpensive and OER active transition metals [2]. However, the exact catalytic mechanism of spinels is still unknown and been actively studied.

In our work, we deposited epitaxial thin films of NiFe₂O₄ inverse spinel on Nb:STO (100), (110) and (111) oriented substrates via PLD and studied with several methods of analysis. With RHEED and XRD we demonstrated the crystallinity and the growth orientation of the epitaxial films both in the in- and out-of-plane directions. All the samples were studied with XPS and AFM both before and after OER electrochemical cycling to demonstrate changes in the spectra and morphology, respectively. In our work we found a clear difference in the OER activity of the samples, which depended on the orientation of the film. Our results are the first step in developing the design rules for spinel-based catalysts.

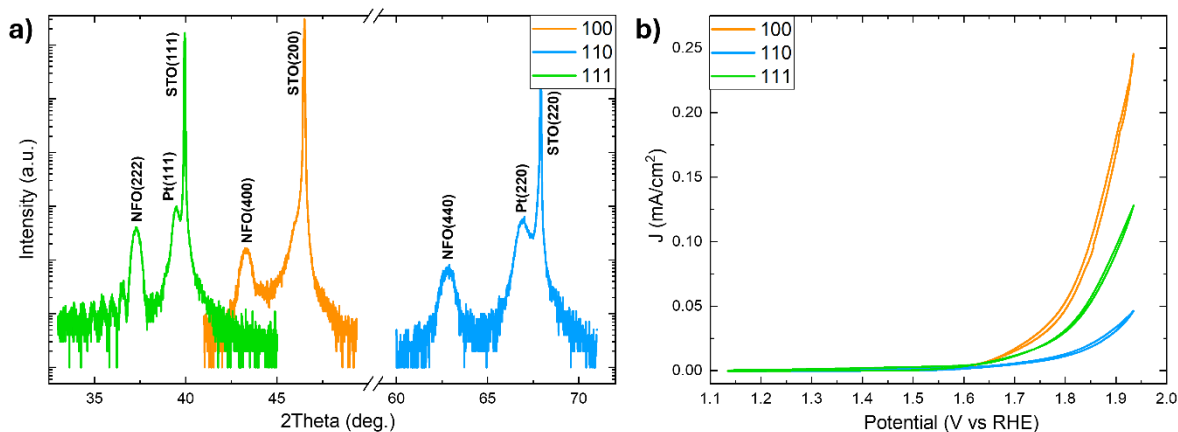


Figure 5. a) XRD of the differently oriented NiFe₂O₄ epitaxial films on Nb:STO(100), (110) and (111). b) 15th cycle of the cyclic voltammetry, performed on the same samples.

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Drug delivery efficiency of polymer-modified iron oxide nanoparticles loaded with chrysin

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Drug delivery systems offer promising solutions for minimizing side effects associated with traditional cancer therapies. Iron oxide nanoparticles (IONPs) are advantageous due to their high binding efficiency and potential for magnetically targeted drug delivery. However, unmodified IONPs are thermodynamically unstable, prone to agglomeration due to magnetic dipole-dipole attraction and van der Waals forces, leading to recognition by the reticuloendothelial system (RES) and subsequent elimination from circulation [1, 2]. Coating strategies with long-chain polymers such as polyethylene glycol (PEG), dextran (DEX), and chitosan enhance stability in ionic environments and facilitate drug adsorption.

This study focuses on synthesizing and characterizing iron oxide nanoparticles (Fe_3O_4 , diameter ~20-25 nm) coated with PEG, DEX, and chitosan, loaded with the natural anticancer bioflavonoid chrysin. Chrysin (5,7-dihydroxyflavone) exhibits potent anticarcinogenic, anti-inflammatory, and antioxidant properties.

Characterization techniques including scanning electron microscopy (SEM) for morphology and size assessment, X-ray diffraction (XRD) for phase analysis using $\text{Cu K}\alpha$ radiation, and Fourier-transform infrared spectroscopy (FTIR) for nanoparticle-drug conjugation analysis were employed. The amount of chrysin loaded onto coated magnetic nanoparticles was quantified using UV-Vis spectrometry. This comprehensive approach aims to develop effective drug delivery systems using polymer-coated IONs for targeted cancer therapy.

Key words: iron oxide nanoparticles, polymer-coating, drug loading efficiency, chrysin

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Effect of the concentration and thermal annealing temperature to the physical properties of GO/PVA composite materials

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Activated carbon fibers, activated carbon and CNT, separately PVA and GO have specific capacities of 60, 90, 27, 240 F/g, respectively, while the value of specific capacity of GO/PVA as a composite material is 400 [1]. This means that the supercapacity of GO/PVA-based materials is 92% for 400 cycles. It indicates the possibility of further improving their physical and structural properties by carrying out the concentration of composite materials of this type, as well as the thermal treatment of the materials obtained at each concentration at different temperatures.

Examining how the characterization of devices and elements based on GO/PVA depends on temperature and concentration is crucial. The findings of present work will make it possible to optimize the parameters of the components and devices that are made using these composites. Simultaneously, identifying temperature-resistant materials through sample thermal stability testing and applying them to pertinent fields are important problems.

Our study revealed that there is a fundamental shift in the optical characteristics when the mass concentration of GO nanostructures distributed within PVA is altered. The band gap value of PVA changed when GO nanostructures were added, and these changes were mostly dependent on the GO concentration. Furthermore, samples that had undergone heat treatment showed temperature-dependent changes in the band gap value. Testing the samples' thermal stability at different GO percentages revealed that the stability increased with GO percentage in the PVA polymer matrix.

Structural analysis (XRD), optical properties (UV-Vis), morphology (TEM) and electrical measurements of the synthesized composite materials were performed.

The dielectric constant had the lowest value at room temperature; yet, it generally increased in value as temperature rose. Polar polymers include in PVA. Temperature had an impact on the mobility of polar molecules, which strengthened polarization and increased the dielectric constant's value. The strength of the electric field prevents Brownian motion from causing chaos because of the temperature.

According to the electrical measurement's outcome, annealing at high temperatures can help reduce GO's functional groups, which will boost the material's electrical conductivity and lower its specific resistance. Simultaneously, the GO/PVA composite's structural reorganization can alter temperature and lead to increased GO sheet alignment and connectivity, improving charge routes and lowering resistance. Specific resistance decreases as a result of improved charge carrier mobility and conductivity brought about by the removal of imperfections.

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[P2-48]

Tuning the electric properties of CaTiO₃ by doping and incorporating GO

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CaTiO₃ (CTO) is a naturally occurring compound and serves as the prototype for perovskite materials. CTO offers unparalleled advantages due to its natural abundance, low cost, and non-toxic nature. Its tunable properties, versatile composition, and high lattice symmetry make CTO-based ceramics highly appealing for applications in various fields, such as dielectric ceramics [1], optoelectronics [2], and photocatalysis [3]. Recent research [4] has revealed that donor-doped CTO-based ceramics exhibit promising thermoelectric (TE) performance, positioning them as potential candidates for energy conversion materials. One major bottleneck for CTO as an effective TE material is its extremely low electrical conductivity. In this work, we would like to demonstrate two strategies for significantly enhancing the electrical conductivity of CTO. The first approach is to substitute Ca²⁺ with cations of higher valence states (>2+), thereby significantly enhancing the carrier concentration [5]. The second strategy is to incorporate graphene oxide into CTO-based compounds to enhance carrier mobility [6].

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High frequency ferromagnetic resonance in hard magnetic iron oxides

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Devices based on non-reciprocal magnetic material response can allow full duplex signal processing in the arising Internet of Things (IoT) era. The use of common ferrites in applications like sensors, wireless communications, radars and advanced driver assistance systems is challenging because used frequency bands are pushed towards the 60 - 300 GHz range for applications in 6G wireless transmissions [1], [2]. Spinel ferrites with external field up to 1T present ferromagnetic resonance (FMR) frequencies up to 40 GHz, while harder [Sr,Ba]-hexaferrites can cover 20-70 GHz. The use of strong external magnetic fields using permanent magnets is not desirable for devices as it increases weight, size and cost. This could be reduced using hard magnetic ferrite oxides with very strong uniaxial anisotropy, like Sr-Ca-Al hexaferrite [3] and epsilon iron oxide (ϵ - $A_x\text{Fe}_{2-x}\text{O}_3$) [4]. Their higher natural FMR frequencies (above 100 GHz) may allow 6G non-reciprocal devices without or with small external magnetic field. We explore the frequency range from 1 to 220 GHz exploiting standard CPW, quasi-optical methods and probe station based experimental set-ups to assess the high frequency FMR response of sub-micron sized powder samples prepared by chemical methods. All experimental set-ups use a vector network analyzer directly or with frequency extenders for 90-220 GHz range, to measure the absorption spectrum versus frequency. We compare our results with reference samples measured by quasi optical spectroscopy. While ϵ - Fe_2O_3 nanoparticles show a narrow FMR around 175 GHz and [Sr,Ca][Al,Fe] $_{12}\text{O}_{19}$ hexaferrite shows it at 125 GHz, FMR in the 1-67 GHz range are found in materials such as oriented single crystals of GaFeO_3 and reference micron sized $\text{BaFe}_{12}\text{O}_{19}$ hexaferrite powders. We can confirm the high frequency FMR in the different magnetic ferrite oxides and correlate them to structure and magnetic properties. The probe station approach with the possibility of applying in-situ magnetic fields may enable the exploration of FMR-assisted magnetization reversal in thin films, nanoparticles or epitaxial layers, as promising candidate for ultra-fast magneto electronic devices [5].

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Spin-torque ferromagnetic resonance in SrTiO₃-based systems

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Spin-orbit torques (SOTs) offer a promising approach for fast, low-power magnetization manipulation in information technologies. Two-dimensional electron gases (2DEGs) formed at oxide interfaces are known to exhibit high SOT efficiency, attracting significant interest for efficient SOT devices [1]. While SOTs in oxide 2DEGs have been observed in various studies using spin-torque ferromagnetic resonance (ST-FMR), one of the most widely used techniques for evaluating SOT efficiency, a recent study revealed a strong artifact inherent in ST-FMR measurements for oxide-based devices [2]. The commonly used oxide substrates for hosting the 2DEGs, such as SrTiO₃ and KTaO₃, act as capacitors due to their high dielectric constant. Therefore, in a ferromagnetic metal(FM)/oxide-substrate device, the resistor of the FM layer R_{FM} and the capacitor formed in the oxide substrate $C_{\text{substrate}}$ are connected in parallel as shown in Fig. 1(a). As a result, part of the radio frequency (RF) current will flow through the oxide substrate at the frequency of the ST-FMR measurement ($f = 5 - 12$ GHz). The Oersted field H_{Oe} due to the leakage RF current in the oxide substrate $I_{\text{substrate}}$ induces a torque acting on the FM layer. This substrate-induced torque produces an artifact ST-FMR signal which possesses the same symmetry as that of SOT (Fig.1 (b)), and thus can strongly affect the evaluation of the SOT efficiency.

In this study, we have investigated SOTs in various SrTiO₃-based devices using ST-FMR. Our results show that the torque efficiency is almost independent of the formation of the 2DEG and scales with the thickness and saturation magnetization of the ferromagnetic layer. This behavior is distinct from conventional SOT mechanisms, indicating the dominant role of the Oersted field generated by off-phased leakage RF currents in the SrTiO₃ substrate. We demonstrate that the efficiency of the substrate-induced torque is comparable to the SOT efficiency of Pt, an archetypal spin-to-charge converter, highlighting the significant impact of this artifact. These findings prompt a re-evaluation of previous experimental findings and the understanding of SOTs in oxide two-dimensional electron gases to advance the design of high-efficiency spintronic devices.

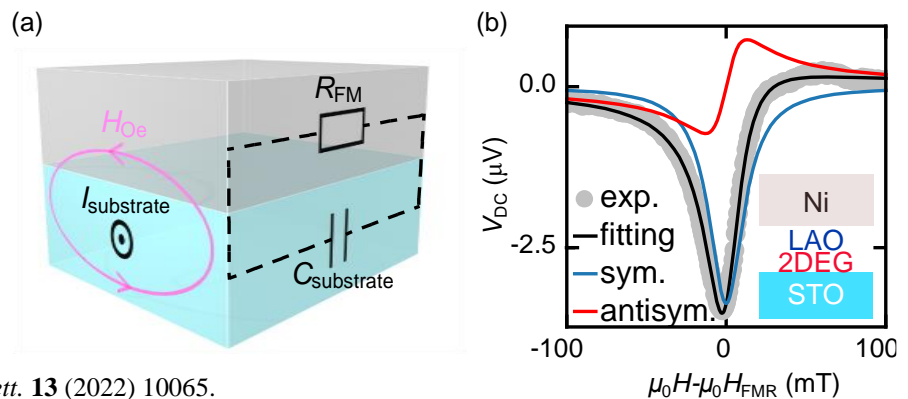


Fig.1 (a) A schematic illustration of a FM/oxide-substrate device for the ST-FMR measurements. (b) A ST-FMR spectrum of the SiO₂(4 nm)/Ni(7 nm)/LAO(5 uc)/STO device.

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[P2-51]

Strong field-driven suppression of the thermoelectric conversion in magnetic oxides

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Suppression of the thermoelectric conversion by the spin Seebeck effect (SSE) upon application of large magnetic fields is a result of the Zeeman-interaction, which induces an energy shift in the magnon dispersion and a reduction of the magnon population of the system [1].

Here, we will show that upon increasing the magnetic compensation of a ferrimagnet a larger magnetic-field-driven SSE suppression can be observed, at least three times larger than for conventional $\text{Y}_3\text{Fe}_5\text{O}_{12}$ system. By evaluation of the magnon-polaron SSE we can estimate the magnon dispersion [2], and discuss the results in terms of possible contributions from magnons with opposite spin polarization and an increased magnon relaxation.

These results highlight the capability of the SSE as an efficient tool to study the spin dynamics and, particularly, the magnon-polaron SSE to obtain information of the magnon dispersion characteristics in magnetic systems.

References

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