

Poster Session A

(Sessions 1, 2, 3, 7, 8)

Date & Time : September 28, 2026 (Mon) 14:00 - 16:00

Venue : Main Hall / Poster Area

Universal Four-Fold Symmetry in Infinite-Layer Nickelates: From the Insulating Parent to the Superconducting State

Muntaser Naamneh¹, Eran Maniv¹, Harold Y. Hwang², and Eytan Grosfeld¹

¹ *Department of Physics, Ben-Gurion University of the Negev, Beer-Sheva, 84105, Israel*

² *Department of Applied Physics, Stanford University, Stanford, California 94305, USA*

The discovery of superconductivity in nickelates has motivated intensive efforts to identify the fundamental interactions that mirror or distinguish them from high T_c cuprate superconductors. Nickelate superconductors, which are hole-doped within the layered structure of $RNiO_2$, share structural similarities with high T_c cuprate superconductors. However, despite similarities in formal valence and crystal symmetry, the fundamental nature of the superconducting state and the parent compound phase in nickelates remains elusive. Strong electronic correlations in infinite-layer nickelates suggest a potentially complex phase diagram, akin to that observed in cuprates, yet a key question about the magnetic ground state remains unanswered. Through magnetoresistance measurements, we observe a universal fourfold rotational symmetry that persists across the entire doping range, from the weakly insulating parent compound to the superconducting state. Notably, this four-fold symmetry displays a $\pi/4$ phase shift with doping or applied magnetic field. This universal symmetry and its characteristic phase shift strikingly parallel the behavior of the electron-doped cuprates, where such signatures are tied to underlying magnetic order. Our modeling suggests that this persistent anisotropy is consistent with the influence of an underlying antiferromagnetic order, providing a unified framework for the electronic ground state throughout the nickelate phase diagram.

Polarization-Dependent EXAFS Analysis of Infinite-Layer Nickelate $\text{Nd}_{1-x}\text{Eu}_x\text{NiO}_2$

Youngdo Kim¹, Dung Vu¹, Sangjae Lee¹, Frederick J. Walker¹, Charles H. Ahn^{1,2,3}

¹ Department of Applied Physics, Yale University, New Haven, CT, USA

² Department of Physics, Yale University, New Haven, CT, USA

³ Department of Mechanical Engineering and Materials Science, Yale University, New Haven, CT, USA

The recently discovered infinite-layer nickelate superconductor $\text{Nd}_{1-x}\text{Eu}_x\text{NiO}_2$ (NENO) has drawn significant attention for its exotic properties, such as field re-entrant superconductivity driven by interactions with magnetic Eu ions [1]. Formed by the removal of apical oxygen atoms along the c-axis from its 3D perovskite precursor, NENO exhibits a strong 2D structural anisotropy distinct from its parent phase [2]. Focusing on this structure anisotropy, we performed polarization-dependent Extended X-ray Absorption Fine Structure (EXAFS) measurements at the Ni K-edge and fitted the EXAFS data using geometric scattering paths corresponding to each polarization direction. The results reveal the presence of additional scattering paths beyond those expected for an ideal 2D infinite-layer structure. Specifically, the out-of-plane EXAFS spectra shows clear scattering signals from apical oxygens, indicating the presence of a residual 3D perovskite phase. Furthermore, the inclusion of the Ni-Ni scattering path from metallic Ni was essential for an accurate fit of the experimental data [3]. Our study demonstrates that polarization-dependent EXAFS is a powerful tool for quantitatively analyzing structural distortions and mixed phases that emerge during the reduction process of infinite-layer nickelates.

WITHDRAWN

References

- [1] D. Vu et al., *Nat. Commun.* **17**, 70254 (2026).
- [2] W. Wei et al., *Sci. Adv.* **9**, eadh3327 (2023).
- [3] J. Gainza et al., *Chem. Mater.* **37**, 4729 (2025).

Epitaxial Growth and Electronic Properties of Layered Nickelate $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$

Hideyuki Kawasoko¹, Kana Nishimura², Kenta Natsui¹, Soichi Kikkawa¹, Seiji Yamazoe¹,
Tomoteru Fukumura²

¹ Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University, Japan

² Department of Chemistry, Graduate School of Science, Tohoku University, Japan

Perovskite-type nickelates have been extensively studied, as exemplified by the metal-insulator transition observed in RENiO_3 (RE = rare-earth elements) [1]. Recently, we reported that in a polycrystalline bulk sample of layered nickelate $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ (Fig. 1a), composed of rock-salt-type $\text{Sr}_{1.5}\text{Bi}_{0.5}\text{O}_2$ layers and perovskite-type SrNiO_3 layers, control of the Sr/Bi ordering of in the $\text{Sr}_{1.5}\text{Bi}_{0.5}\text{O}_2$ layers leads to a reduction of electrical resistivity by approximately two orders of magnitude in the ordered $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ phase compared to the disordered $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ phase [2]. Thin film growth of $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ is of particular interest for further exploration of the physical properties. However, to date, there have been no reports on the growth of $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ thin films. In this study, we grew $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films via solid-phase epitaxy and evaluate their electronic properties.

Precursor films were deposited at room temperature on $\text{LaAlO}_3(001)$ single crystalline substrates by RF magnetron sputtering using a cation-stoichiometric target prepared from SrCO_3 , Bi_2O_3 , and NiO . The deposited precursor films were subsequently heated in air at 1000 °C to grow $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ thin films via solid-phase epitaxy.

X-ray diffraction measurements revealed that the $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ thin films were oriented along [001] direction (Fig. 1b). A small amount of NiO impurity was also detected. Additionally, the presence of 101 asymmetric reflections confirmed the ordered arrangement of Sr/Bi cations. Reciprocal space mapping indicated that the films were epitaxially grown on the LaAlO_3 (001) substrate. The lattice constants of the $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films were determined to be $a = 5.346$ Å and $c = 17.72$ Å, indicating compressive strain compared to those of polycrystalline bulk $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ ($a = 5.377$ Å, $c = 17.49$ Å). In contrast to the polycrystalline bulk $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$, which exhibits metallic temperature dependence, the $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films showed a high electrical resistivity and semiconducting temperature dependence despite their high crystallinity (Fig. 1c). This behavior is likely attributable to epitaxial strain and/or the presence of NiO impurity phase.

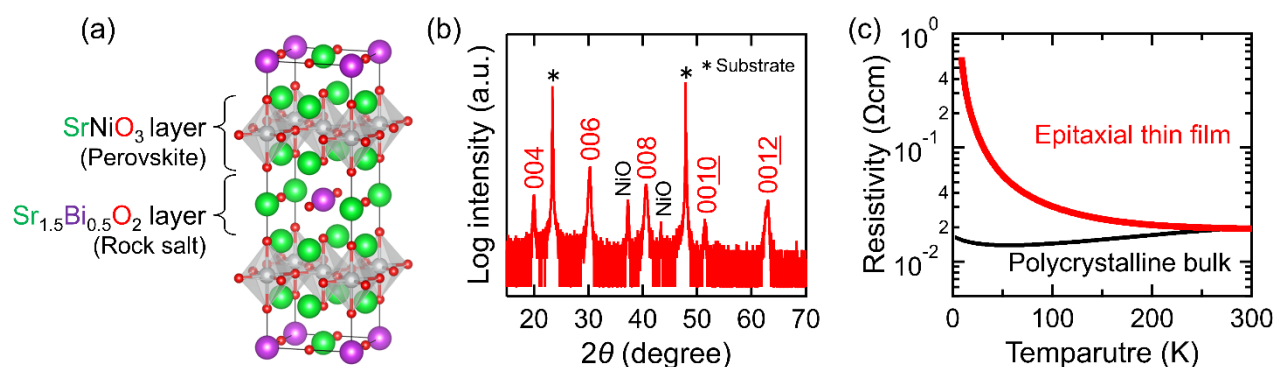


Figure 1 (a) Crystal structure of $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$. (b) X-ray diffraction pattern and (c) temperature dependence of electrical resistivity for $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films.

References

- [1] M. Imada *et al.*, Rev. Mod. Phys. **70**, 1039 (1998).
- [2] K. Matsumoto, H. Kawasoko *et al.*, Adv. Sci. **10**, 2304978 (2023).

Superconductivity in As-Grown Bilayer Nickelate Thin Films

Rahaf Yaseen¹, Gideok Kim¹, Michael Neumann¹, Eva Benckiser¹, Moritz Karcher¹,
Bernhard Keimer¹

¹ *Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany*

The discovery of superconductivity in bilayer nickelates has brought an important new perspective to the research field of unconventional superconductivity in strongly correlated oxide materials. To date two material systems have been identified to host superconductivity: infinite-layer Nickelates and bilayer Ruddlesden-Popper (RP) $\text{La}_3\text{Ni}_2\text{O}_7$ [1, 2]. While superconductivity in bulk $\text{La}_3\text{Ni}_2\text{O}_7$ was initially observed under high pressure, recent studies have demonstrated superconducting behavior in epitaxial thin films at ambient pressure [3, 4]. Despite this progress, the synthesis of superconducting bilayer nickelate thin films remains challenging because of the metastable nature of the Ruddlesden-Popper phase and the stringent requirements on stoichiometry, strain, and oxygen content.

In this work, we investigate chemical substitution as a route to stabilize superconductivity in bilayer nickelate thin films. Ca and Sm substituted $\text{La}_3\text{Ni}_2\text{O}_7$ thin films were synthesized by ozone-assisted molecular beam epitaxy (MBE) on $\text{LaSrAlO}_4(001)$ substrates. Remarkably, superconducting behavior is observed directly in the as-grown films without the low-temperature post-annealing treatments commonly employed to induce superconductivity in bilayer nickelates.

A superconducting $\text{La}_{2.85}\text{Ca}_{0.15}\text{Ni}_2\text{O}_7$ thin film was successfully obtained in the as-grown state. X-ray diffraction measurements indicate a highly ordered crystal structure and full epitaxy with the underlying substrate. Electrical transport measurements reveal a superconducting transition near 45K. The superconducting properties remain stable under ambient conditions for several weeks and can be further preserved by storage in liquid nitrogen.

In addition, a series of $\text{La}_{3-x}\text{Sm}_x\text{Ni}_2\text{O}_7$ thin films ($x=0.6-1.2$) was synthesized. Structural characterization reveals a systematic evolution of the crystal structure with increasing Sm concentration. Electrical transport measurements demonstrate a strong dependence of the electronic properties on Sm substitution. All Sm-substituted samples exhibit either partial or full superconducting transitions in the as-grown state without additional thermal treatments, with the most pronounced transition observed for $x=1.2$ near 25K.

These results demonstrate that chemical substitution and ozone-assisted MBE provide an effective pathway for stabilizing superconducting behavior in bilayer nickelates thin films.

References :

- [1] G. Zhou et al., *Nature* **640**, 641 (2025)
- [2] H. Sun et al., *Nature* **621**, 493 (2023)
- [3] E. K. Ko et al., *Nature* **638**, 935 (2025)
- [4] B. Hao et al., *Nature Mater.* **24**, 1756 (2025)

Evolution in Ruddlesden-Popper nickelate via soft-chemistry

Antoine Devincenti¹, Michal Kiaba¹, Jennifer Fowlie¹

¹ Materials Science & Engineering, Northwestern University, Evanston, Illinois, United States

While significant progress has been made in synthesizing superconducting Ruddlesden-Popper (RP) phases as thin films, further control over their evolution and stabilization is needed to enable more in-depth study and understanding. It has been reported that an ozone atmosphere, during or after growth, is crucial to stabilize superconductivity in RP nickelates. Given the numerous polymorphs of RP, fine control of the growth conditions is crucial to ensure selective and high crystalline quality. In addition to the continuous improvement of RP nickelate films deposited via pulsed laser deposition, several routes toward film stabilization are investigated. Some of our experiments have shown that different annealing routes have led to various states of RP and even to transformations of the crystal structure. Using high-temperature X-ray diffraction (HTXRD), we probed the soft chemical evolution of an ozone-annealed RP thin film. The nature of this RP crystal-structure evolution remains debated. Some reports suggest the formation of a higher-order RP phase, while others suggest the intercalation of oxygen into the lanthanum oxide rock-salt layer [1, 2]. Preliminary results on the reversibility of this crystalline evolution will be discussed.

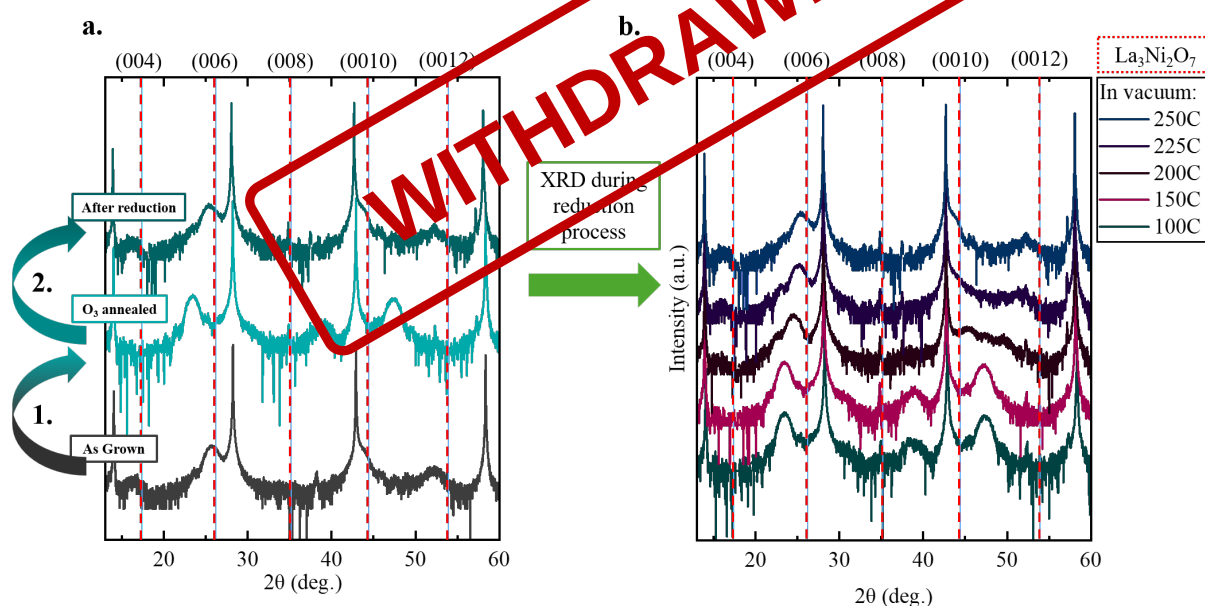


Figure 1: **a.** Evolution of the Ruddlesden-Popper structure with ozone annealing (arrow 1). Evolution of ozone annealed crystal under reducing environment (arrow 2) **b.** Intermediate states of the crystal structure acquired during the reduction process (**a.** arrow 2).

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Epitaxial Growth of La-Site-Substituted Bilayer Nickelate Thin Films and Emergent In-Plane Octahedral Rotation

Lee Kihyuk¹, Sihun Seong¹, Seo Hyoung Chang¹, Bongju Kim¹, Minu Kim¹

¹ Department of Physics, Chung-Ang University, Seoul, Republic of Korea

Recent studies have reported that applying high pressure (~ 15 GPa) to bulk $\text{La}_3\text{Ni}_2\text{O}_7$ leads to high T_c (~ 80 K) [1], and that a similar effect can be achieved in thin films through epitaxial strain engineering via lattice mismatch ($T_c \sim 40$ K) [2]. These findings have motivated extensive research on structural tuning as a means of increasing T_c . Here, we explore site-selective substitution of smaller-radius cations (Pr, Sm) at the La site as an additional structural tuning parameter, in addition to epitaxial strain. Despite the narrow growth window, bilayer nickelate thin films were successfully grown by pulsed laser deposition (PLD) on LaSrAlO_4 (LSAO) substrates, and confirmed to be epitaxially strained to the substrate by transport measurements and X-ray scattering including reciprocal space mapping (RSM). While the La and Pr films retain the strain-imposed structure, we find that Sm substitution drives the emergence of an in-plane octahedral rotation absent under epitaxial strain alone. A-site chemical pressure thus activates an in-plane structural degree of freedom, which modifies the in-plane $\text{Ni } 3d_{x^2-y^2} - \text{O } 2p$ hybridization, adding an in-plane axis to the structure-superconductivity relationship of $\text{La}_3\text{Ni}_2\text{O}_7$.

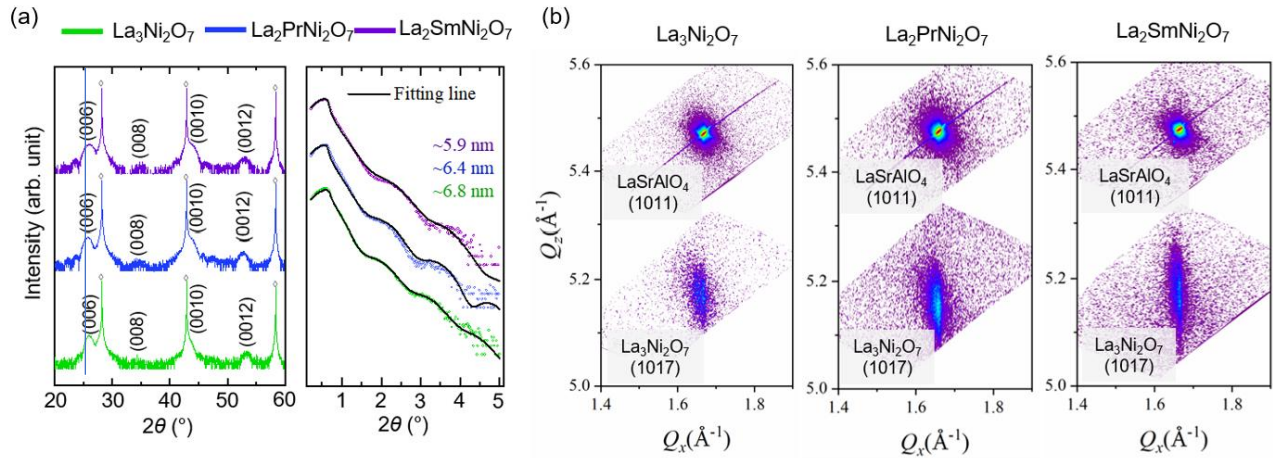


Figure 1: (a) X-ray diffraction (XRD) and X-ray reflectivity (XRR) confirming the phase-pure growth of the bilayer nickelate thin film. (b) Reciprocal space map (RSM) revealing the epitaxial relationship and in-plane strain state of the film with respect to the substrate.

References

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Synthesis and investigation of Eu_2NiO_4 in Ruddlesden-Popper structure

Hao-Bo Li^{1*}, Daichi Ito², Shunsuke Kobayashi², Yuki Sasahara³, Takuya Majima^{4,5}, Yoshihiro Tsujimoto⁶, Akira Chikamatsu⁷, Hyoungjeen Jeon⁸, Hiroshi Takatsu³, Hiroshi Kageyama³, Hidekazu Tanaka^{1*}

¹*SANKEN, The University of Osaka, Ibaraki, 567-0047, Japan.*

²*Nanostructures Research Laboratory, Japan Fine Ceramics Center, Nagoya, 456-8587 Japan.*

³*Department of Energy and Hydrocarbon Chemistry, Graduate School of Engineering, Kyoto University, Kyoto 615-8510, Japan.*

⁴*Quantum Science and Engineering Center, Kyoto University, Uji 611-0011, Japan.*

⁵*Department of Nuclear Engineering, Kyoto University, Kyoto 615-8540, Japan.*

⁶*National Institute for Materials Science, Tsukuba Ibaraki Namiki 1-1 305-0044 Japan.*

⁷*Department of Chemistry, Faculty of Science, Ochanomizu University, 2-1-1 Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan.*

⁸*Department of Physics, Pusan National University, Busan 46241, Korea.*

The strongly correlated oxides with perovskite structure have attracted vast attention due to the intrinsic complex interactions between different freedoms. Though nickel oxide with Ni^{1+} ($3d^9$) has long been considered as an analog to Cu-based superconducting material, it is only since 2019 the superconductivity and high- T_c have been discovered in perovskite nickel oxide^{1,2}. Then much effort has been put to the synthesis of nickel oxides with perovskite and their derived structure. Especially, high quality single crystal samples with delicately controlled composition and strain are highly required for the research in this area. By aiming to further explore the potential nickel-based superconductors, we have challenged the materials synthesis and the characterization of unreported Eu_2NiO_4 with Ruddlesden-Popper structure. In this work, we report the first Eu_2NiO_4 thin film that is successfully grown on LaAlO_3 substrate with pulsed laser deposition. The scanning transmission electron microscopy provides clear evidence of alternatively stacked EuNiO_3 and EuO layers, which agrees well with $n = 1$ Ruddlesden-Popper structure. By introducing Sr as dopant, a metal-insulator transition is observed. Our work provides not only an unreported material but also an emergent platform for research in strongly correlated systems.

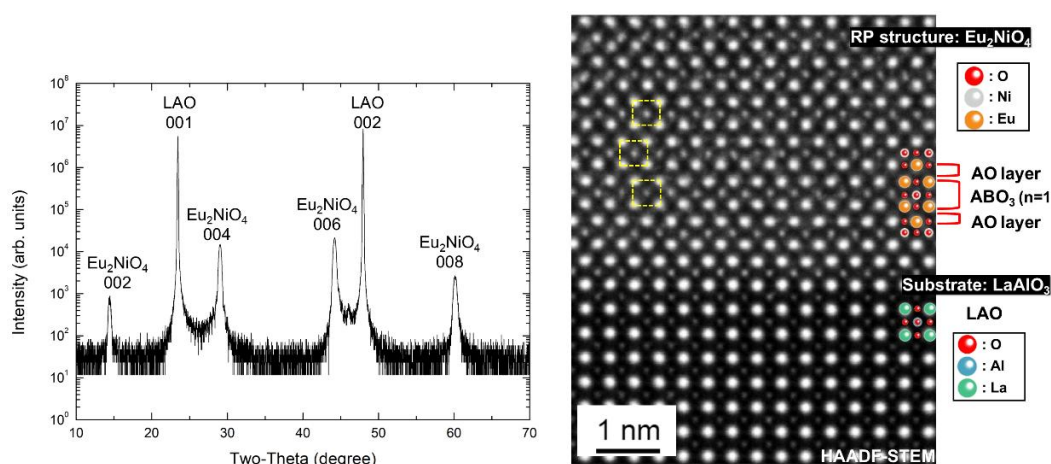


Figure 1. XRD pattern of Eu_2NiO_4 and the cross-section STEM image near the interface.

References

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Metallic Conduction in $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ Epitaxial Thin Films by Controlling Growth Condition

Kenta Natsui¹, Hideyuki Kawasoko¹, Soichi Kikkawa¹, Seiji Yamazoe¹

¹*Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University, Japan*

Transition metal oxides have been extensively studied due to various interesting physical properties [1]. Among them, $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$, which is a layered nickelate composed of rock-salt $\text{Sr}_{1.5}\text{Bi}_{0.5}\text{O}_2$ layers and perovskite SrNiO_3 layers, exhibits a large variation in electrical resistivity by controlling order and disorder of the Sr/Bi arrangement in the rock-salt layers [2], attributed to modulated hybridization between Ni 3d and O 2p orbitals [3]. While this variation has been observed only in bulk polycrystals, extending it to thin-film form is of great interest for device applications. To date, films of the ordered $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ phase were reported to be grown via solid phase epitaxy, but they exhibited semiconducting behavior [4], in contrast to the metallic behavior of ordered bulk polycrystals [3]. In this study, we grew $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films by solid-phase epitaxy using rf-magnetron sputtering with heating rate ranging from 20 to 1000 °C/min. In addition to the mixed-oxide target ($\text{SrCO}_3/\text{Bi}_2\text{O}_3/\text{NiO}$) used in the previous study, we also used a single-phase target composed of $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$.

X-ray diffraction measurements confirmed the epitaxial growth of the ordered $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ films on $\text{LaAlO}_3(001)$ substrates, grown from both the mixed-oxide and single-phase targets, hereafter referred to as the mixed-oxide-target films and single-phase-target films, respectively. The crystallinity of both mixed-oxide-target and single-phase-target films improved with increasing heating rate. While the mixed-oxide-target films contained NiO impurity, the formation of NiO impurity was significantly suppressed in the single-phase-target films (Fig. 1a). As a result, the single-phase-target films exhibited metallic conduction, displaying a clear metallic temperature dependence of electrical resistivity (Fig. 1b). In this presentation, we will discuss the origin of the metallic conduction observed in this study, including electronic structure analysis based on X-ray absorption fine structure measurements.

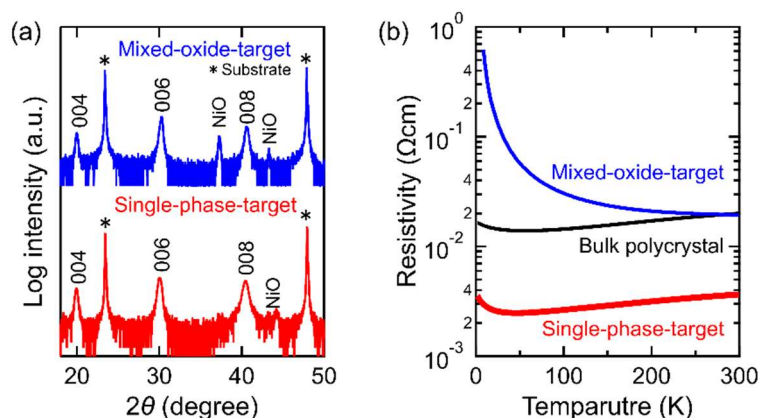


Figure. 1 (a) X-ray diffraction patterns and (b) temperature dependence of electrical resistivity for $\text{Sr}_{2.5}\text{Bi}_{0.5}\text{NiO}_5$ epitaxial thin films.

References

- [1] M. Imada *et al.*, Rev. Mod. Phys. **70**, 1039 (1998).
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- [4] K. Nishimura *et al.*, in preparation.

Thickness Dependence of Electronic Structure and V-V Dimerization in VO₂ (110)_R Ultrathin Films

S. Inoue¹, D. Shiga^{1,2}, R. Hayasaka¹, K. Ozawa², and H. Kumigashira^{1,2}

¹ IMRAM, Tohoku University, Sendai, Japan

² Photon Factory, IMSS, KEK, Tsukuba, Japan

VO₂ is a promising candidate as a channel material for future Mott transistors since it exhibits an abrupt metal-insulator transition (MIT) near room temperature accompanied by V-V dimerization. Recent studies have demonstrated that the properties of VO₂ ultrathin channel layers vary depending on the orientation of the dimerization axis (one-dimensional V-V chains) along the [001]_R direction relative to the confinement (growth) direction defined by the surface and interface [1–3]. Thus, for designing VO₂ channel layers with tailored performance, it is crucial to obtain information on the dimerization-orientation-dependent characteristics of nanostructured VO₂. Here, we investigated changes in the electronic and crystal (characteristic V-V dimer) structures of thickness-controlled VO₂ (110)_R films, in which the dimerization axis is perpendicular to the confinement direction, by *in situ* photoemission spectroscopy (PES) and x-ray absorption spectroscopy (XAS), respectively.

Figure 1(a) shows the thickness (t) dependence of the *in situ* valence-band spectra for VO₂ (110)_R ultrathin films measured at 420 and 300 K. For the thick 20-nm films ($T_{\text{MIT}} \sim 360$ K), the spectral changes characteristic of the temperature-driven MIT in VO₂ [4] are observed. The line shapes of V 3d states near the Fermi level (E_F) remain almost intact even when t is reduced down to 4.5 nm, suggesting the invariance of the properties of VO₂ (110)_R. On the other hand, below 4.5 nm, the density of states at E_F for the 420-K spectra is steeply reduced and eventually disappears at 1 nm, forming a Mott gap [1]. A Fermi-edge profile exists down to 1.5 nm, indicating the occurrence of a thickness-dependent MIT at a critical thickness of 1–1.5 nm. To further investigate the insulating state stabilized below the critical thickness (two-dimensional limit) in terms of V-V dimerization, we performed polarization-dependent O K-edge XAS. As shown in Fig. 1(b), an additional peak appears around 530.6 eV, which is observed only in the monoclinic insulating phase and can be identified with the $d_{\parallel}^*$ state, which is a fingerprint of V-V dimerization in monoclinic VO₂ [5]. Indeed, the $d_{\parallel}^*$ state is clearly observed in the thick 20-nm spectra. With decreasing t , the $d_{\parallel}^*$ peak begins to weaken and eventually disappears below the critical thickness, indicating that V-V dimerization no longer occurs for VO₂ (110)_R films in the two-dimensional limit. Thus, the PES and XAS results indicate that VO₂ (110)_R in the two-dimensional limit, in which the characteristic V-V dimerization axis is perpendicular to the confinement direction, exhibits a Mott insulating nature without V-V dimerization, as a result of the dominance of Mott instability.

References

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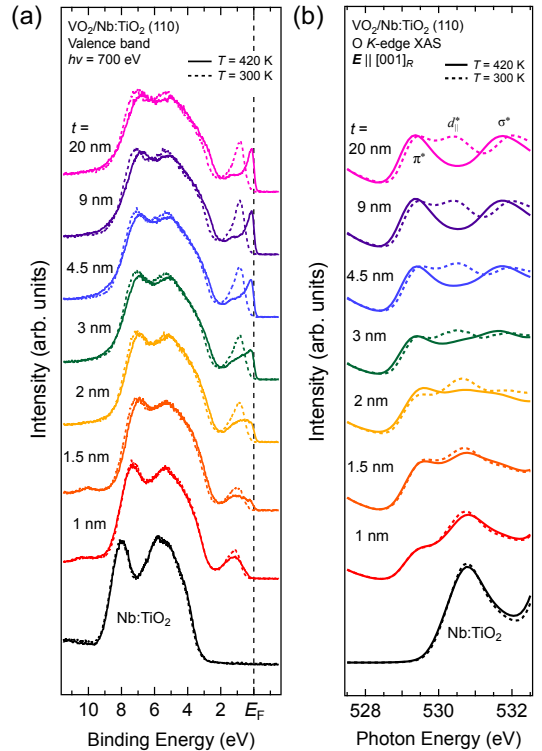


Figure 1: Thickness dependence of *in situ* (a) valence-band and (b) O K-edge XAS spectra measured at 420 and 300 K for VO₂/Nb:TiO₂ (110) films.

Thickness-Periodic Mott Insulators in CaRuO₃ Ultrathin Films: An Extraordinary Size Effect in Correlated Oxides

Masahito Sakoda¹, Hiroyoshi Nobukane², Shuhei Shimoda³, Koichi Ichimura¹

¹ Department of Applied Physics, Graduate School of Engineering, Hokkaido University

² Department of Physics, Graduate School of Science

³ Institute for Catalysis, Hokkaido University

Strongly correlated oxides provide an ideal platform for creating artificial low-dimensional quantum phases, because their electronic states can be strongly modified by lattice, dimensionality, and boundary conditions. We have investigated CaRuO₃ ultrathin films as an artificial two-dimensional electron system in which a perovskite ruthenate located near a quantum critical region is confined to a thickness comparable to the Fermi wavelength. High-quality CaRuO₃ ultrathin films were grown on NdGaO₃ (110) substrates by molecular beam epitaxy with atomically flat interfaces. We found extraordinary alternating metal-insulator transitions depending on film thickness with a period of approximately 25 Å. The electrical resistivity changes by three orders of magnitude at room temperature and by nine orders of magnitude at low temperature, far exceeding conventional quantum size effects [1]. The same periodicity was confirmed by ultraviolet photoelectron spectroscopy during in situ etching, and the anisotropy between (110) and (001) growth indicates that the effect is governed by the electronic structure rather than by simple quantum confinement [1].

We further clarified that the metal-insulator transition can be controlled by growth parameters, including the Ca/Ru molecular flux ratio and substrate temperature, through lattice expansion toward the Mott-insulating phase [2]. Calcium-rich growth conditions, especially around Ca/Ru $\approx$ 1.6 with a flat surface roughness of approximately 1.4 Å, enhance the extraordinary size effect and reproduce thickness periods of 23.4–26.3 Å [3]. Recently, in-plane X-ray diffraction revealed a 24.8 Å oscillation of the lattice spacing, synchronized with the insulating thicknesses. Magnetotransport and Hall measurements showed reduced carrier density and antiferromagnetic correlations in the expanded lattice state. A nesting vector of the square-pole Fermi surface produces a spin-density wave with a real-space period of 23.5–27.6 Å along the [110] direction, matching the observed size effect [4]. These results demonstrate that nanoscale boundary conditions select a spin-density-wave standing wave, which triggers periodic Mott localization in CaRuO₃. Thickness therefore acts as a new control parameter for quantum phase transitions in correlated oxide electronics.

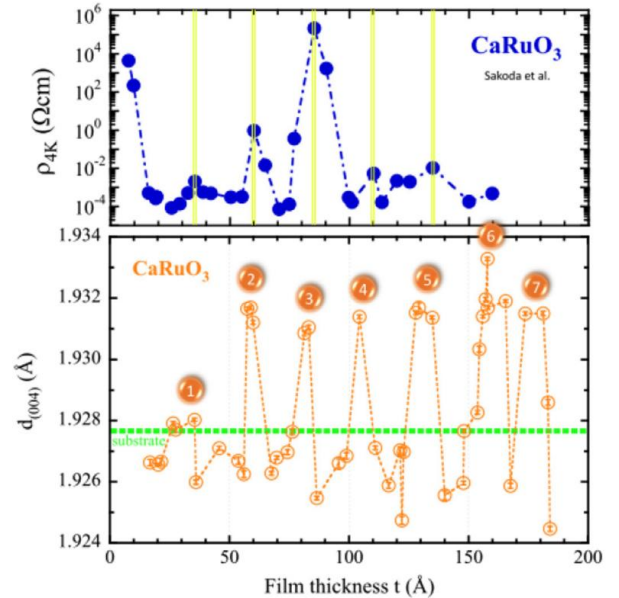


Figure 1: Thickness-periodic Mott transition in CaRuO₃ ultrathin films, showing electrical-resistivity oscillation, and lattice expansion.

References

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Carrier-Doping-Induced Metal to Insulator Crossover in CaRuO₃ Thin Films

Y.Hamada¹, N.Obara¹, R.Yoshino¹, H.Nobukane²,
S.Shimoda³, M.Sakoda¹

¹*Dept. of Appl. Phys. Hokkaido Univ.*, ²*Dept. of Phys. Hokkaido Univ*

³*Institute for Catalysis. Hokkaido Univ.*

CaRuO₃, a 4d transition-metal oxide, is known as a strongly correlated oxide and is located in the quantum critical region of the Mott–Hubbard phase diagram. It has been reported that when CaRuO₃ thin films are epitaxially grown along the (110) direction on NdGaO₃ substrates, they exhibit a thickness-dependent periodic metal–insulator transition[1]. From the values of the activation energy and ARPES measurements, this transition was identified as a Mott transition driven by a spin-density wave[2].

In this study, we investigated whether the above mentioned Mott transition could be induced by electrostatic carrier doping in CaRuO₃ thin films. Carrier doping was achieved by applying a gate voltage to the samples through an electric double-layer transistor (EDLT). In addition, the thin films were fabricated by molecular beam epitaxy (MBE) to obtain high-quality surface conditions. Prior to the measurements, the ionic liquid was degassed, allowing the application of higher gate voltages. Low-temperature measurements were then performed in-situ. To account for the substantial thickness-dependent changes in electrical conduction, transport measurements were carried out on CaRuO₃ thin films with a wide range of thicknesses. Gate-voltage-induced insulating behavior was observed shown in Figure1. In addition to the temperature dependence of the resistivity, magnetoresistance and other transport measurements will be presented to provide further insight into the origin of the insulating state.

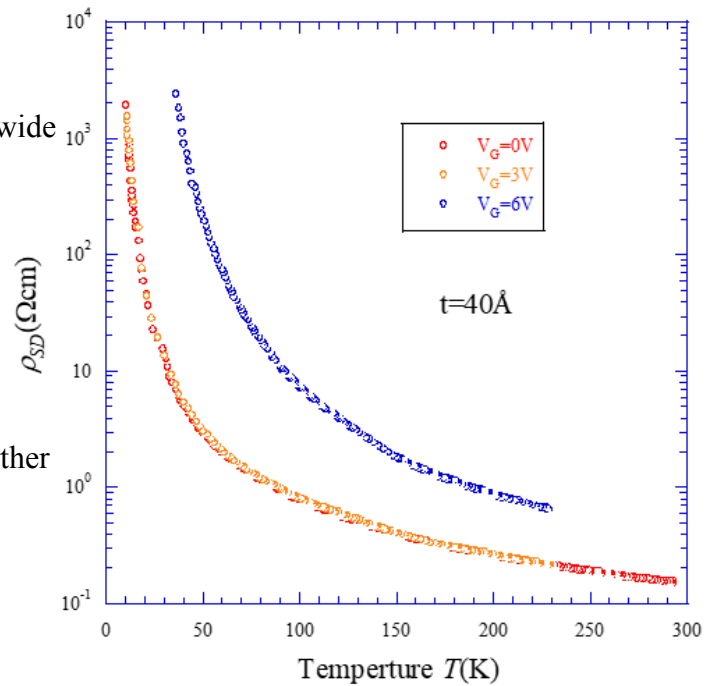


Figure1: Gate-voltage-dependent resistivity behavior.

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Thermal Runaway Instability Drives Electric Field Induced Breakdown of a Mott Insulator

Aniket Bajaj,¹ Anudeep Tullibilli,² Pavan Nukala,² and Bhavtosh Bansal¹

¹*Department of Physical Sciences, Indian Institute of Science Education and Research Kolkata, Mohanpur, West Bengal, 741246, India.*

²*Centre for Nanoscience and Engineering, Indian Institute of Science, Bengaluru 560012, India.*

Electrical excitation of strongly correlated materials exhibiting an insulator-to-metal transition (IMT) enables ultrafast and highly scalable resistive switching, making them promising candidates for futuristic computing paradigms such as neuromorphic computing. However, the fundamental mechanism underlying this switching remains controversial due to presence of thermally induced metal-insulator-transition (MIT). To address this, we investigate the physical origin of electric field induced breakdown in a well-known strongly correlated material, vanadium dioxide (VO_2) using simultaneously performed transport and optical imaging measurements. We establish that the observed resistive switching in such materials arises from a thermal runaway instability driven by the material's pronounced negative temperature coefficient of resistance. We demonstrate that this instability emerges exclusively under voltage-driven excitation, where positive thermal feedback causes uncontrolled Joule heating and the formation of highly localized filamentary conduction channels (see figure).

Through an analytical treatment of the macroscopic heat equation, we derive the exact physical criterion dictating the onset of this runaway regime, which is validated by direct experimental observations. By mapping the transition dynamics onto the normal form of saddle-node bifurcation theory, we reveal that the abrupt transition corresponds to the annihilation of steady-state thermodynamic equilibrium solutions. Moreover, we present that the reset mechanism is governed by a complementary pinch-off instability of the filament, driving the abrupt metal-to-insulator transition. Beyond advancing non-equilibrium transport physics and clarifying the precise thermodynamic and nonlinear dynamical origins of this switching furnishes the essential framework required to harness IMT materials for next-generation artificial computing paradigms.

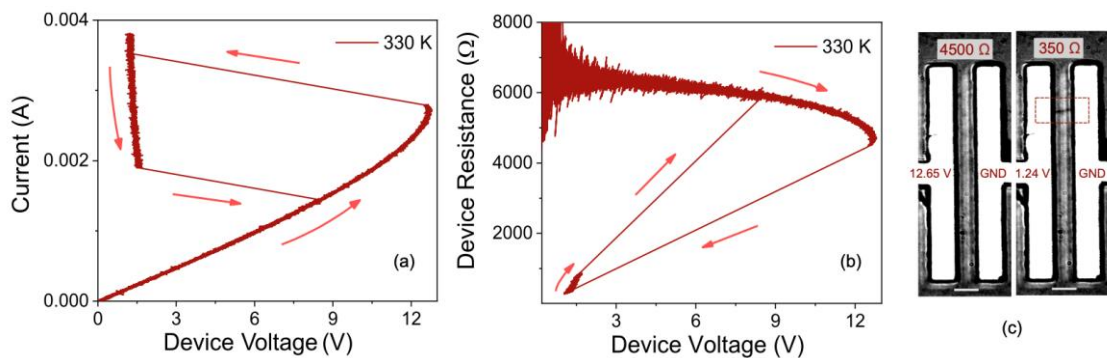


Figure: (a) Current-voltage characteristics obtained under a voltage ramp, showing an initial linear regime followed by a pronounced nonlinear response with abrupt IMT and on ramping down the voltage, system undergoes abrupt MIT. (b) Corresponding device resistance as a function of device voltage. (c) Optical images of the device channel before and after breakdown, showing the formation of a localized conductive filament.

Solid-phase synthesis of Cr-doped V_2O_3 thin films on $\alpha\text{-Al}_2\text{O}_3$ (0112) substrates exhibiting metal-insulator transition at room temperature

Josei Fujiwara¹, Akira Ohtomo¹, Kohei Yoshimatsu¹

¹ *Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan*

V_2O_3 exhibits two phase transitions: from a low-temperature antiferromagnetic insulating (AFI) phase to a paramagnetic metallic (PM) one at ~ 150 K (AFI-PM transition), and from the PM phase to a paramagnetic insulating (PI) one at ~ 500 K (PM-PI transition) [1]. The corundum-type crystal symmetry remains intact across the PM-PI transition, and its temperature is shifted close to room temperature by $\sim 1\%$ Cr doping. These characteristics make Cr-doped V_2O_3 a promising material for application in oxide electronics. However, the PM-PI transition disappears in a thin-film form, presumably due to lattice strain introduced during non-equilibrium thin-film growth.

We thus applied reactive solid-phase synthesis combined with a crystalline seed layer to Cr-doped V_2O_3 thin films exhibiting the PM-PI transition. Cr-doped V_2O_3 seed layers with ~ 10 nm thickness was first grown on $\alpha\text{-Al}_2\text{O}_3$ (0112) substrates at 800°C using pulsed-laser deposition with a 0.5% Cr-doped V_2O_3 polycrystalline ceramic target. An amorphous precursor Cr-doped V_2O_3 film was subsequently deposited at room temperature and annealed at 800°C in an H_2 atmosphere, resulting in the formation of crystalline 0.5% Cr-doped V_2O_3 films (~ 300 nm thickness).

Out-of-plane X-ray diffraction profiles (Fig. 1) revealed that the seed layer promoted crystallization of the precursor films with the same out-of-plane orientation as the substrate, although an additional 3030 reflection was observed. Temperature-dependent resistivity measurements (Fig. 2) showed not only the low-temperature AFI-PM transition but also the PM-PI transition with an approximately doubled resistivity near room temperature. These results indicate that solid-phase synthesis with the crystalline seed layer on $\alpha\text{-Al}_2\text{O}_3$ (0112) substrates is effective for realizing Cr-doped V_2O_3 films exhibiting the clear PM-PI transition.

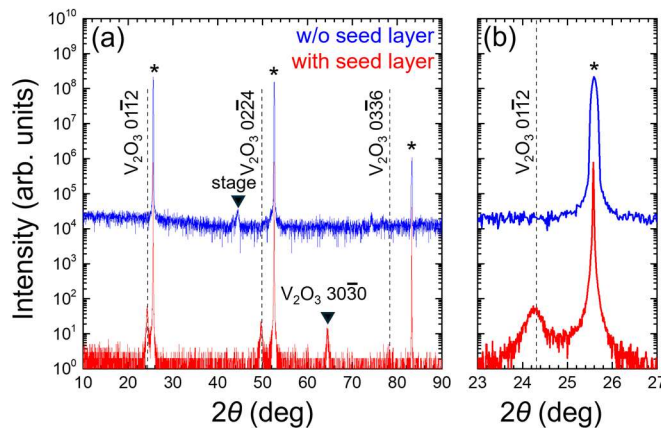


Fig. 1. Out-of-plane X-ray diffraction profiles of Cr-doped V_2O_3 films on $\alpha\text{-Al}_2\text{O}_3$ (0112) substrates (a) in the wide 2θ range and (b) around the V_2O_3 0112 reflection. Asterisks indicate reflections from $\alpha\text{-Al}_2\text{O}_3$ substrates.

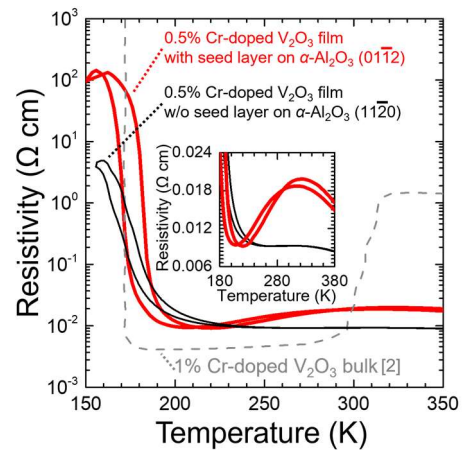


Fig. 2. Temperature-dependent resistivity for the Cr-doped V_2O_3 film with the seed layer. The inset shows magnification of the ρ - T curve near the PM-PI transition.

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Theoretical Study of SVILC Qubits in Bi-2212 Cuprate Thin Films

Yuto Takatsu¹ and Hiroyasu Koizumi²

¹ Degree Programs in Pure and Applied Sciences, University of Tsukuba

² Center for Computational Sciences, University of Tsukuba

Our group has been proposing a theory for cuprate superconductivity in which superconducting loop currents, “spin-vortex-induced loop currents (SVILCs)” are produced in the CuO_2 plane [1]. Each SVILC is characterized by a topologically protected winding number, and this degree of freedom is expected to be applicable to the construction of qubits. Bi-2212 cuprate thin films provide a promising platform for studying such SVILC-based quantum states and their possible application to quantum devices.

In our model, a nano-island composed of four SVILCs forms a qubit. Different current patterns in a single nano-island give rise to 16 qubit state candidates. As shown in Fig. 1, many of their energy levels depend on the externally applied current, and the 16 states can be classified into four groups according to their energy-level structure.

To extend this idea to multi-qubit systems, we investigate coupled nano-islands in which coupling currents are applied through quantum-dot sites placed between neighboring nano-islands, as schematically shown in Fig. 2. The dependence of the energy-level structure on external and coupling currents is evaluated by numerical simulations. We also discuss how the coupling between nano-islands can modify the energy-level structure and induce controlled transitions among selected current-pattern states. This study extends our recent work on SVILC qubit systems [2] and provides a theoretical basis for exploring quantum gate operations in cuprate-based quantum devices.

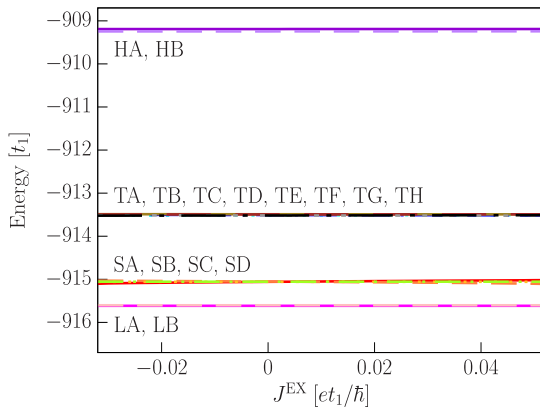


Figure 1: Energy levels of the sixteen current-pattern states in a single nano-island as a function of the externally applied current. The states are classified into four groups according to their energy-level structure. Parameters: $t_1 = 200$ meV, $et_1/\hbar = 4.87 \times 10^{-5}$ A.

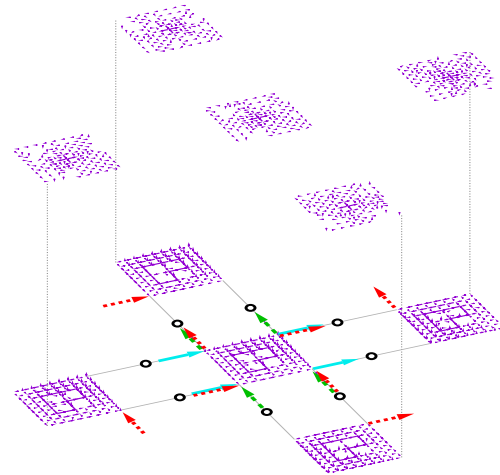


Figure 2: Schematic of coupled nano-islands in a Bi-2212 cuprate thin film. Open circles denote quantum-dot sites connecting neighboring nano-islands. Cyan solid and green dashed arrows indicate externally applied currents along the x- and y-directions, respectively, while red dotted arrows indicate coupling currents between nano-islands. Purple vectors denote current vectors in the CuO_2 planes.

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Revealing the temperature-dependent phase transitions of LaTaO₄ in Carpy-Galy structure

Yongjin Shin¹

¹*Department of Semiconductor Convergence Engineering, Dankook University, Yongin-si 16890, Republic of Korea*

LaTaO₄ is a Carpy-Galy type layered perovskite ($A_nB_nO_{3n+2}$) well established as a geometrically induced ferroelectric, yet realizing its ferroelectric functionality remains challenging. The stable ground state is a monoclinic nonpolar structure, while the known orthorhombic ferroelectric phase is metastable and appears only above 600 K [1]. The phase transition between the two structures further passes through an incommensurate regime whose structural solution has not been clarified, leaving the microscopic mechanism yet to be understood. Using first-principles calculations, this work identifies a previously unknown polar monoclinic phase that is lower in energy than the orthorhombic ferroelectric structure. Group-theoretical analysis reveals that the coupling of specific phonon modes creates an energy barrier along the nonpolar-to-polar pathway, providing a natural explanation for the observed transition hysteresis and capturing the key structural features of the incommensurate regime. Finally, we survey how this energy barrier evolves with chemical substitution, identifying opportunities to tune the ferroelectric energetics for high-k material applications.

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Magnetic Field Induced Superconductivity in NbO

Sandra Glotzer,^{1,2} Elena Williams,¹ Jeong Rae Kim,^{1,2} Tommaso Chiarotti^{1,2},
David Abramovitch^{1,2}, Adrian Llanos,^{1,2} Marco Bernardi,^{1,2} and Joseph Falson^{1,2}

¹ *Department of Applied Physics and Materials Science, California Institute of Technology,
Pasadena, California 91125, USA*

² *Institute for Quantum Information and Matter, California Institute of Technology, Pasadena,
California 91125, USA*

Conventionally, an external magnetic field suppresses superconductivity via the paramagnetic effect, which aligns electron spins, and the orbital effect, which raises the kinetic energy of electrons. However, as predicted by Kharitonov and Feigelman [1], a rare interplay between local magnetic moments and a parallel magnetic field can lead to an enhancement of the critical temperature with increasing field. Experimental realizations of this phenomenon have been limited to systems doped with magnetic ions or impurities [2-4]; furthermore, this effect has yet to be observed in an oxide system.

Here, we report the first demonstration of field-induced superconductivity in NbO, a metallic and non-magnetic oxide. Employing ultra-high-quality thin films of NbO [5], we find that the system progresses from field-enhanced to field-induced behavior as dimensionality is reduced. We link our experiments to first-principles theory, which uncovers evidence of magnetization of the Nb-terminated surface arising from localized *d* orbitals. Modelling the spin scattering dynamics of this system within the Kharitonov-Feigelman (KF) framework [1] suggests that the polarization of these local moments by the magnetic field, which suppresses magnetic fluctuations, is responsible for field-induced superconductivity. Our work ushers in oxide systems as a new platform for studying the anomalous enhancement of superconductivity, wherein broken symmetries and correlation effects can play a strong role.

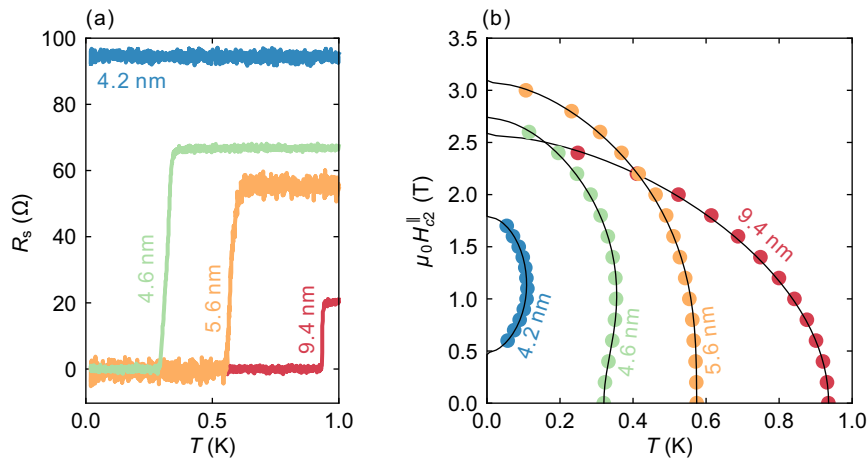


Figure 1: Field-induced superconductivity in NbO. (a) Zero-field temperature-dependent sheet resistance of NbO films with varying thickness. (b) In-plane critical field as a function of temperature. Solid lines are fits to the KF model.

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Anion-Gradient Engineering of Superconductivity in NbO-Based Oxynitride Epitaxial Thin Films

Linshen Xing¹, Yasumichi Cho¹, Kenichi Kaminaga¹, Yuto Abiko¹, Shingo Maruyama^{1,2}, and Yuji Matsumoto¹

¹ Department of Applied Chemistry, Tohoku University, Sendai, 980-8579, Japan

² Department of Engineering, Shizuoka University, Shizuoka, 432-8561, Japan

Rock-salt NbO has recently attracted attention as a superconducting oxide thin-film material, while its superconducting properties are strongly affected by epitaxial strain, especially in films thinner than 100 nm [1]. Given that NbN has long been known as a robust superconductor, similarly having a rock-salt framework, rock-salt niobium oxynitrides Nb(O,N) potentially also exhibit intriguing superconducting properties depending on the composition. In this study, we make the first attempt to further introduce “nanoscale anion-gradient engineering” in fabricating Nb(O,N) epitaxial films, in which NbO-rich and NbN-rich regions are spatially designed along the growth direction, and investigate how this depth-dependent anion profile could modify superconducting transport and diamagnetic response.

Compositionally-graded and homogeneous Nb(O,N) oxynitride films with a nominal thickness of 40 nm were grown on MgO(001) substrates by pulsed laser deposition [2] using NbO and NbN targets. The total thickness was designed as the O/N ratio was controlled along the growth direction to form NbO-derived anion-gradient structures on a nanoscale: G-NbNO with an NbO-rich surface, G-NbON with an NbN-rich surface, and homogeneous H-NbON, all of which films have a nominal average composition of NbO_{0.5}Nb_{0.5}. SIMS, XPS, XRD, and reciprocal-space mapping confirmed that all the films were epitaxially grown with the intended depth-dependent anion distributions.

Among the Nb(O,N) films, G-NbNO exhibited the most favorable superconducting properties, showing a zero-resistance transition temperature of $T_c^{\text{zero}} = 9.4$ K, which is very close to that of a NbN reference film ($T_c^{\text{zero}} = 9.9$ K), together with a comparable sharp resistive transition and the strongest diamagnetic response (Figure 1). Furthermore, its upper critical field reached $\mu_0 H_{c2} = 9.5$ T, exceeding that of the NbN reference film (8.6 T). These results demonstrate that introducing the anion-gradient in Nb(O,N) is an effective approach to tuning its superconductivity with macroscopic superconducting uniformity relatively maintained despite its compositional ununiformity.

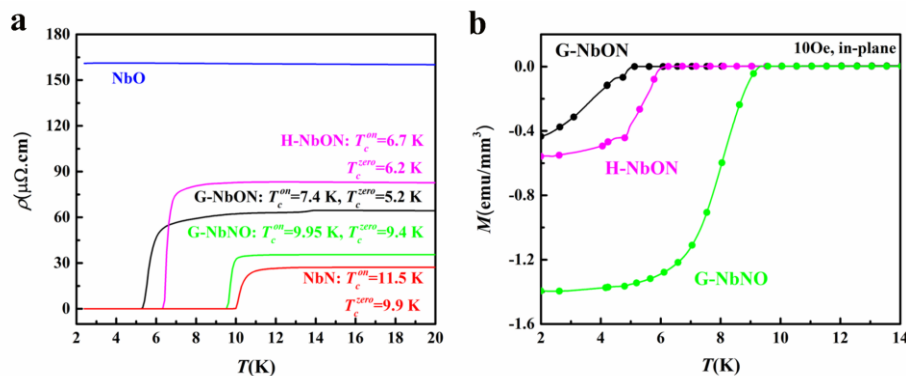


Figure 1: Superconducting transport and diamagnetic response of NbO-based Nb(O,N) epitaxial thin films.
(a) Resistivity–temperature curves of graded and homogeneous Nb(O,N) films with NbO and NbN references.
(b) Magnetization–temperature curves measured under an in-plane field of 10 Oe.

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Repeated temperature modulation epitaxy of superconducting LiTi_2O_4

Shu Takashima¹, Kohei Yoshimatsu¹, Akira Ohtomo¹

¹ *Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan*

LiTi_2O_4 (LTO) is the only superconductor among spinel-type oxides, and its electrical properties vary significantly depending on the x composition in bulk $\text{Li}_{1+x}\text{Ti}_{2-x}\text{O}_4$ [1]. The epitaxial growth of stoichiometric LTO thin films has been attempted using various techniques including pulsed-laser deposition (PLD) and molecular beam epitaxy (MBE). In general, higher growth temperatures (T_g) lead to better crystallinity and superconductivity. However, due to the high volatility of Li species, adjusting the stoichiometric composition becomes difficult at high T_g . To solve this dilemma, we have adapted repeated temperature modulation (RTM) epitaxy with PLD [2], in which repeated growth sequences at high and low T_g satisfy both improved crystallinity and stoichiometry.

The RTM-PLD was implemented for epitaxy of LTO on MgAl_2O_4 (111) substrates under an Ar/H_2 atmosphere by optimizing ranges of T_g , excess Li amounts in targets, and thickness during a single cycle ($T_g = 740\sim 850^\circ\text{C}$, $[\text{Li}]/[\text{Ti}] = 1\sim 1.5$, and ~ 7 nm, respectively). The crystallinity and electrical properties of the obtained films were investigated using X-ray diffraction (XRD) and temperature-dependent resistivity (ρ) measurements and compared with those of a conventional high- T_g sample.

Out-of-plane XRD profiles revealed that both films w/ and w/o RTM were apparently single phases [Fig. 1 (a)] and highly crystallized as evidenced from narrow rocking curve widths [Fig. 1 (a) inset]. However, broad reflection tails were observed only for the film w/o RTM, suggesting the presence of a Li-deficient phase in the film [Fig. 1 (a)]. Temperature-dependent ρ for the film w/ RTM indicated a lower residual ρ than that of one of the best-reported MBE samples [3] [Fig. 1 (b)]. Moreover, the residual ρ ratio (RRR = $\rho_{300\text{ K}}/\rho_{15\text{ K}}$) of the former was 6.67, exceeding that of the latter 5.85 [Fig. 1 (b) inset]. To gain deeper insight, we qualitatively estimated the degree of excess Li (x composition) occupying the B sites by taking the ratios of the XRD intensities of the 222 to 111 reflections (I_{222}/I_{111}), which reflect the difference in effective atomic scattering factors at the B sites [4]. The observed positive correlation between I_{222}/I_{111} and RRR [Fig. 1(c)] clearly demonstrates that reducing the amount of excess Li at the B sites results in improved electrical properties in LTO thin films.

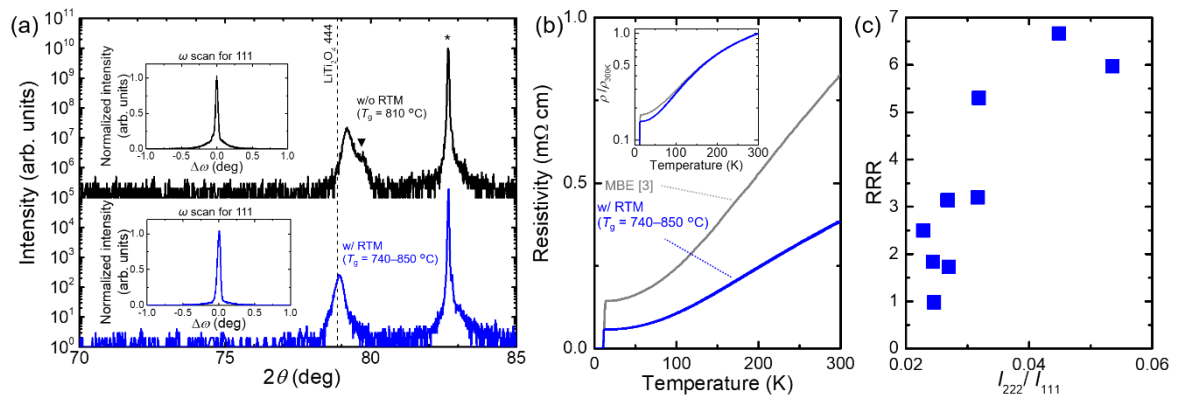


Fig. 1. (a) Out-of-plane XRD profiles of LTO thin films around the LTO 444 reflection. The asterisk and triangle indicate reflections from the substrates and degraded film, respectively. Insets show the rocking curve profiles for the thin-film 111 reflections. (b) Temperature-dependent resistivity of LTO thin films grown w/ and w/o RTM. Inset shows temperature-dependent $\rho/\rho_{300\text{ K}}$. (c) RRR vs. I_{222}/I_{111} for a number of films grown by RTM-PLD.

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Waterfall-like Incoherent States and Electron–Plasmon Coupling in Compressively Strained SrNbO_3 Thin Films

Pampa Sadhukhan¹, Deepnarayan Biswas², Jieyi Liu², Tien-Lin Lee², Lenart Dudy³, Merit Spring¹, Arnold M. Müller^{4,5}, Michael Sing¹, and Ralph Claessen¹

¹ *Physikalisches Institut and Würzburg–Dresden Cluster of Excellence ctd.qmat, Universität Würzburg, 97074 Würzburg, Germany*

² *Diamond Light Source Ltd., Didcot, United Kingdom*

³ *SOLEIL Synchrotron, 91190 Saint-Aubin, France*

⁴ *Laboratory of Ion Beam Physics, ETH Zurich, 8093 Zurich, Switzerland*

⁵ *Ionplus AG, Dietikon, Switzerland*

Recently, 4d transition-metal oxides have emerged as an interesting platform for studying the interplay between non-trivial topology and many-body interactions, as they combine moderate electron correlations with appreciable spin-orbit coupling.

Here, we investigate the electronic structure of compressively strained SrNbO_3 (SNO) thin films, a proposed Dirac semimetal candidate, using soft X-ray angle-resolved (SX-ARPES) and Nb *L*-edge resonant photoemission spectroscopy (ResPES). In addition to the well-known low-energy quasiparticle (QP) band, we observe a previously unresolved waterfall-like in-gap (IG) state (s. Fig. 1). Its occurrence as a high-energy tail of the dispersing QP band – rather than uniformly throughout momentum space – rules out a defect-related origin and demonstrates its intrinsic character.

ResPES measurements show a simultaneous enhancement of the QP and IG spectral weights across the Nb *L*-edge, establishing a common Nb 4d orbital character. To clarify the origin of the incoherent state, we systematically tune the carrier density and correlation strength through varying oxygen stoichiometry and film thickness. The waterfall-like feature is strongest in metallic films and becomes suppressed in more insulating ultrathin films, accompanied by an energy shift from approximately 1.9 eV to 1.0 eV with decreasing carrier density. Consistently, oxygen-rich films exhibit also a red shift of the satellite feature and overall reduced spectral weight. These observations demonstrate that the IG weight is strongly linked to the metallic nature of the SNO films, while electron-electron correlations play only a minor role. Furthermore, analogous satellite structures are observed in the Sr 3d, O 1s, and Nb 3d core-level spectra. Similar plasmon-induced satellite features have recently been identified in SrVO_3 – the more strongly correlated 3d analogue of SNO – by high-resolution ARPES [6]. Our results identify the waterfall-like IG state as a manifestation of electron–plasmon coupling and provide the first spectroscopic evidence of plasmon-induced incoherent states in SNO, establish electron–plasmon coupling as a key ingredient of its low-energy electronic structure.

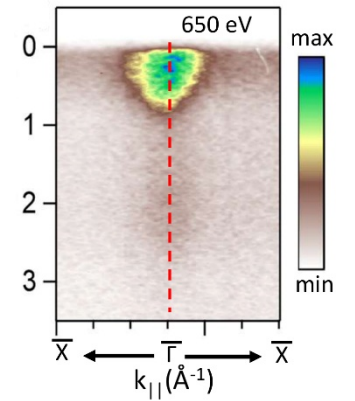


Figure 1: ARPES intensity map, showing the low-energy QP band and waterfall-like spectral IG-weight, consistent with a plasmon-induced satellite state

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Imaging the itinerant-to-localized transmutation of electrons across the metal-to-insulator transition

Andrés F. Santander-Syro¹

¹ *Institut des Sciences Moléculaires d'Orsay, Université Paris-Saclay, Orsay, France*

The most familiar physical property of materials is their ability to conduct (metals) or not (insulators) electric current. The conductivity of metals increases when the temperature decreases. Thus, copper conducts ten times better the current at 70 K than at 600 K. But for V_2O_3 , a metal at room temperature, the conductivity drops sharply by a factor of a million when the temperature falls below 160 K. It becomes a so-called Mott insulator. This metal-to-insulator transition is the paradigmatic, and still unsolved, example of strongly correlated electron physics! How to explain this astonishing transition?

This work imaged for the first time how the itinerant, wave-like electrons in the metallic phase of V_2O_3 localize across the Mott metal-insulator transition [1]. We will discuss the observed changes in electronic structure and compare them with other paradigmatic examples of metal-insulator transitions in correlated oxides.

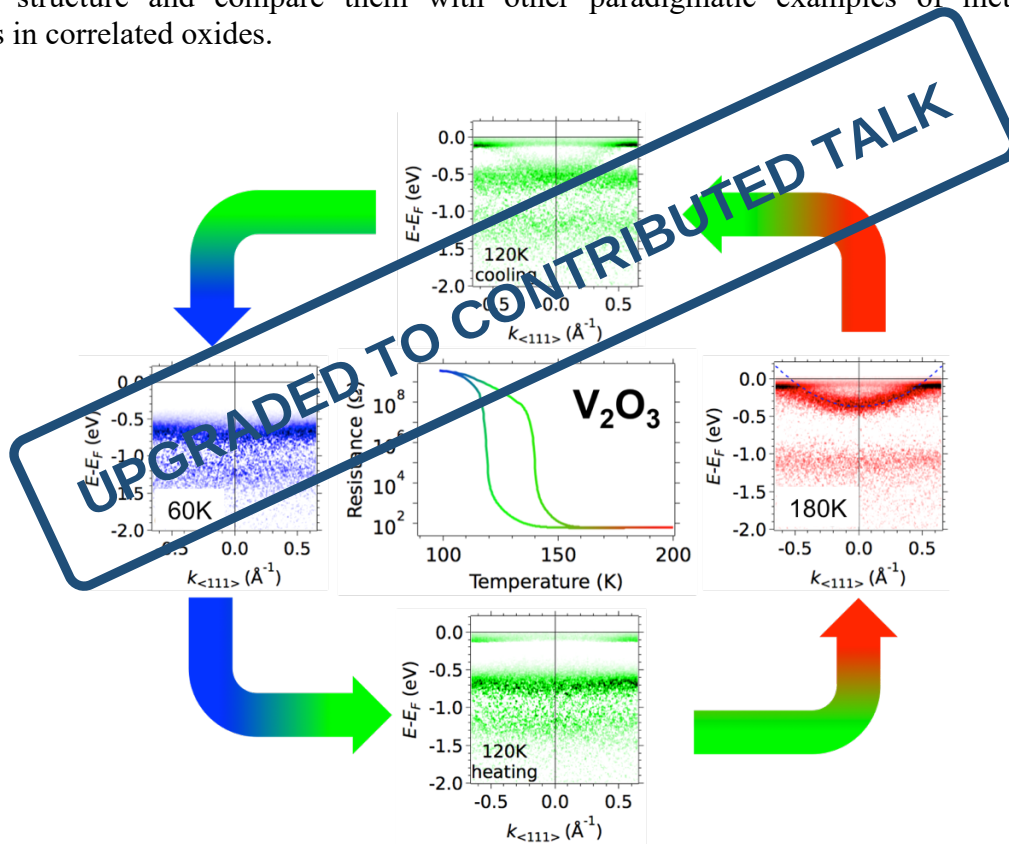


Figure 1: Changes in the electronic structure across the metal-to-insulator transition in V_2O_3 , measured using angle-resolved photoemission spectroscopy.

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Direct Observation of Double Dirac-Cone Crossings and Strain-Enhanced Nontrivial Band Gap in a Topological Crystalline Insulator Thin Film

Takuya Takashiro^{1,2}, Gauthier Krizman³, Mahdi Hajlaoui², Tetiana Zakusylo², Ondrej Caha⁴, Lakshmi Sajeev⁴, Marcin Rosmus⁵, Natalia Olszowska⁵, Dawid Wutke⁵, Gunther Springholz²

¹*Department of Physics, Graduate School of Science, Tohoku Univ.*

²*Institute of Semiconductor and Solid State Physics, Johannes Kepler Univ.*

³*Laboratoire de Physique de l'Ecole normale supérieure, ENS, Univ. PSL, CNRS, Sorbonne Univ.*

⁴*Department of Condensed Matter Physics, Masaryk Univ.*

⁵*National Synchrotron Radiation Centre SOLARIS, Jagiellonian Univ.*

Topological crystalline insulators (TCIs) have attracted considerable interest as their nontrivial band topology is protected by crystalline mirror symmetry, rather than the time-reversal symmetry in Z_2 topological insulators, giving rise to multiple helical Dirac cones in the Brillouin zone. Particularly, $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$, a prototypical TCI material, also exhibits ferroelectric behavior, providing an ideal platform for exploring the interplay between band topology and structural distortion. We previously grew n -type (111)-oriented $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ films with at least 0.25% Bi doping, resulting in direct observation of their Dirac cones by angle-resolved photoemission spectroscopy (ARPES) [1]. However, the polar nature of the (111) surface induces strong surface band bending, causing an unintentional Rashba effect that hinders the systematic investigation of the TCI nature in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$.

To elucidate intrinsic topological properties in ARPES measurements, we focus on nonpolar (001)-oriented $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ thin films. The thin-film samples were grown on KCl (001) substrates with PbSe (001) buffer layers by molecular beam epitaxy. To tune E_F into the conduction-band side, we induce an appropriate surface band bending by depositing 0.3 ML Pb atoms onto the surface instead of heavy Bi doping. As shown in Fig. 1(a) (for $x = 0.52$), we observe double Dirac-cone crossings, characteristic of the TCI (001) surface, in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ for the first time. From the ARPES data, we obtain the nontrivial band gap ($2\Delta < 0$) as a function of temperature, as shown in Fig. 1(b), estimating the nontrivial transition temperature at 225 K. In addition, $|2\Delta|$ in the present case (57 meV at 10 K) is larger than that of a bulk $\text{Pb}_{0.54}\text{Sn}_{0.46}\text{Te}$ system (30 meV at 4.5 K) measured by magneto-optical measurements [2], which indicates that a compressive strain induced by the KCl substrate enlarges the negative-gap magnitude.

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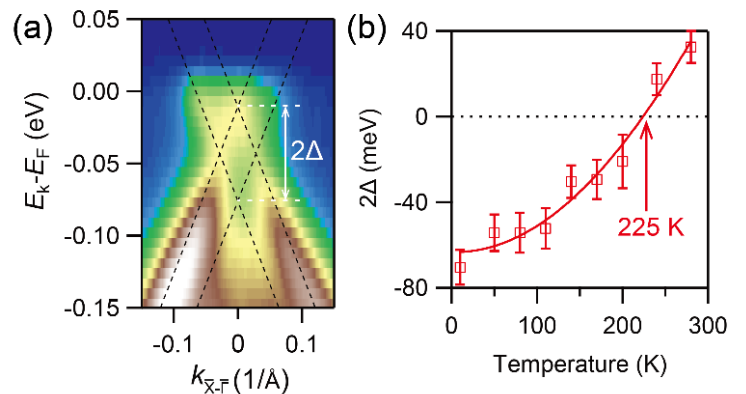


Fig. 1 ARPES spectrum of double Dirac cones in a $\text{Pb}_{0.48}\text{Sn}_{0.52}\text{Te}$ (001) thin film grown on a PbSe/KCl (001) substrate after 0.3 ML Pb deposition. (b) Temperature dependence of the nontrivial band gap (2Δ) of the $\text{Pb}_{0.48}\text{Sn}_{0.52}\text{Te}$ thin film.

Carrier-density-driven evolution of two-dimensional electron systems on β -Ga₂O₃ surface

Ryu Yukawa^{1,*}, Hiroshi Mizuseki², Suryo Santoso Putro³, Yé-Jin L. Lee^{1,4}, Yuuki Masutake³, Hinako Telengut³, Boxuan Li³, Hajime Yamamoto³, Tadashi Abukawa^{1,3}, Junya Yoshida¹, Vladimir V. Kochurikhin^{5,6}, Taketoshi Tomida^{5,6,7}, Masanori Kitahara^{5,6,7}, Takahiko Horiai^{6,8}, Akira Yoshikawa^{5,6,7,8}, Nobuhiko Sarukura^{8,9}, Noriko Chikumoto⁹, Toshihiko Shimizu^{8,9}, Marilou Cadatal-Raduban^{9,10}, Yoshiyuki Kawazoe^{8,11,12}, Ryuhei Kohno¹, Hiroshi Kumigashira³, Takuto Nakamura^{13,14}, Tatsuhiko Kanda¹⁵, Akira Yasui¹⁵, Akihiro Koide¹⁵, Miho Kitamura¹⁶, Hideaki Iwasawa^{16,17,18}, Koji Horiba¹⁶, Kenichi Ozawa¹⁹

¹SRIS, Tohoku University, Japan, ²KIST, Republic of Korea, ³IMRAM, Tohoku University, Japan, ⁴Faculté des Sciences et Ingénierie, Sorbonne Université, France, ⁵IMR, Tohoku University, Japan, ⁶C&A Corporation, Japan, ⁷FOX Corporation, Japan, ⁸NICHe, Tohoku University, Japan, ⁹Institute of Laser Engineering, Osaka University, Japan, ¹⁰Unitec Institute of Technology, New Zealand, ¹¹Department of Physics, SRM University-AP, India, ¹²Institute of Science and Center of Excellence in Advanced Functional Materials, Suranaree University of Technology, Thailand, ¹³Graduate School of Frontier Biosciences, Osaka University, Japan, ¹⁴Graduate School of Science, Osaka University, Japan, ¹⁵JASRI, Japan, ¹⁶NanoTerasu Center, QST, Japan, ¹⁷Synchrotron Radiation Research Center, QST, Japan, ¹⁸Research Institute for Synchrotron Radiation Science, Hiroshima University, Japan, ¹⁹IMSS, KEK, Japan

β -Ga₂O₃ is an ultrawide-bandgap semiconductor regarded as a promising next-generation high-power material. However, the behavior of two-dimensional electron systems (2DESs) formed at its surfaces and interfaces remains largely unexplored. Here, we use angle-resolved photoemission spectroscopy (ARPES) to resolve the evolution of electron-doped β -Ga₂O₃ surface 2DESs as a function of sheet carrier density (n_{2D}). At low n_{2D} (Fig. 1(a)), spectral weight appears near the Fermi level (E_F) together with broad features extending toward higher binding energy (E_B), suggesting polaronic states as reported for oxide 2DESs [1]. With increasing n_{2D} , a well-defined parabolic quasiparticle band develops, indicating a crossover toward a Fermi-liquid-like electronic structure. Notably, the effective mass (m^*) increases with n_{2D} (Fig. 1(b)). This behavior cannot be explained by a simple polaron-to-Fermi-liquid crossover and instead suggests the presence of an additional carrier-density-dependent many-body renormalization mechanism operating in the 2DES at the β -Ga₂O₃ surface [2].

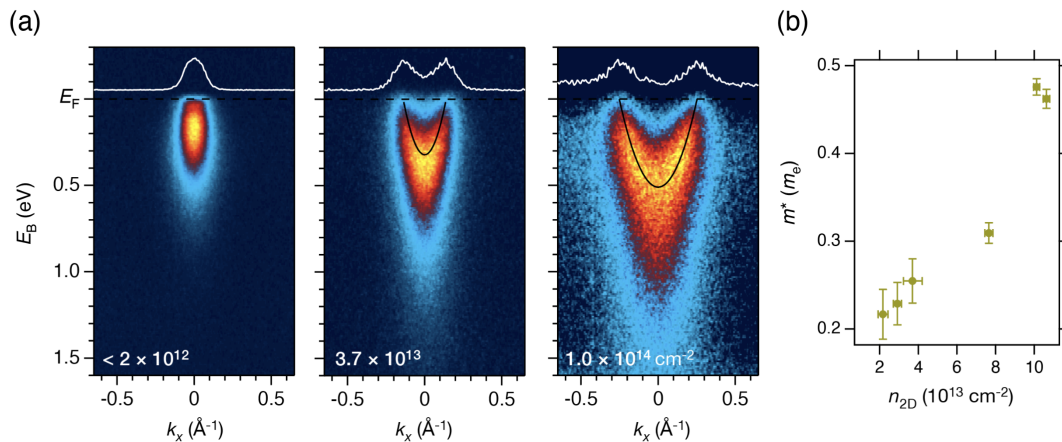


Figure 1: (a) ARPES intensity plots taken at various n_{2D} . (b) Plot of the obtained m^* against n_{2D} .

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Microscopic Origin of Photoinduced Two-Dimensional Electron System Formation at β -Ga₂O₃ Surface

Suryo Santoso Putro¹, Yé-Jin L. Lee^{2,3}, Vladimir V. Kochurikhin^{4,5}, Taketoshi Tomida^{4,5,6}, Masanori Kitahara^{4,5,6}, Takahiko Horiai^{5,7}, Akira Yoshikawa^{4,5,6,7}, Hiroshi Mizuseki⁸, Noriko Chikumoto⁹, Nobuhiko Sarukura^{7,9}, Kenichi Ozawa¹⁰, Daisuke Shiga^{1,10}, Hiroshi Kumigashira^{1,10}, Ryu Yukawa²

¹IMRAM, Tohoku University, Japan, ²SRIS, Tohoku University, Japan, ³Faculté des Sciences et Ingénierie, Sorbonne University, France, ⁴IMR, Tohoku University, Japan, ⁵C&A Corporation, Japan, ⁶FOX Corporation, Japan, ⁷NICHe, Tohoku University, Japan, ⁸Korea Institute of Science and Technology (KIST), Republic of Korea, ⁹Institute of Laser Engineering, The University of Osaka, Japan, ¹⁰IMSS, KEK, Japan.

Formation of a two-dimensional electron system (2DES) on β -Ga₂O₃ is important not only for understanding surface electronic states in ultra-wide-bandgap oxides, but also for exploring electronic properties relevant to future power-semiconductor applications [1]. We recently demonstrated electron doping to high sheet carrier densities exceeding $1 \times 10^{14} \text{ cm}^{-2}$ on β -Ga₂O₃ [2]; however, the microscopic mechanism enabling the formation and control of the resulting 2DES remains unclear.

In this study, the mechanism behind 2DES formation at the Cs-adsorbed β -Ga₂O₃ surface is investigated using angle-resolved photoemission spectroscopy (ARPES) and X-ray photoelectron spectroscopy (XPS) at BL-2A MUSASHI of the Photon Factory, KEK. Cs was deposited *in situ* on the *bc*-plane of β -Ga₂O₃ with estimated coverages of 0.27 and 2.49 atoms/u.c. Near-Fermi-level ARPES spectra and XPS core-level spectra were tracked as a function of synchrotron-light photoirradiation time.

As shown in Fig. 1(a), no 2DES formation is observed on the bare surface, whereas the 2DES emerges on the Cs-adsorbed surface upon synchrotron-light irradiation. This indicates that both Cs deposition and photoirradiation are necessary for the formation of the 2DES. Figures 1(b) and 1(c) show that the Cs 3*d* peak becomes broader after prolonged photoirradiation, indicating that the chemical states of the deposited Cs at the surface change under photoirradiation. Notably, the time required for electron doping and the direction of the Cs peak shift depend on the Cs coverage. These results indicate that 2DES formation is governed by the coupled effects of Cs coverage and photoirradiation-induced chemical-state evolution, providing a microscopic basis for controlling surface electron doping in β -Ga₂O₃.

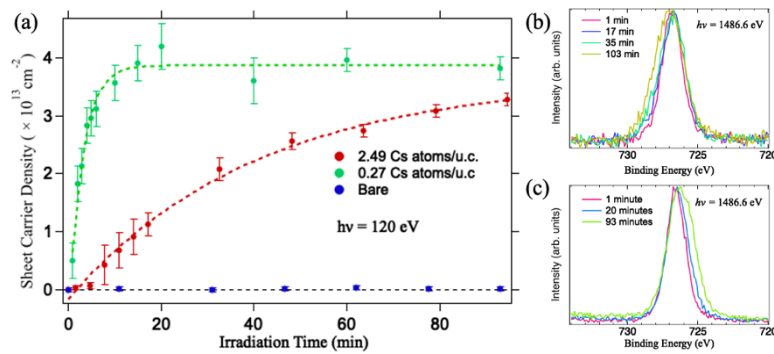


Figure 1: (a) Temporal evolution of the sheet carrier density determined by ARPES for the bare and Cs-adsorbed β -Ga₂O₃ surfaces. (b,c) Temporal evolution of Cs 3*d* XPS spectra for estimated Cs coverage of 0.27 atoms/u.c. (b) and 2.49 atoms/u.c. (c). Here, u.c. denotes the surface 1×1 unit cell of the *bc*-plane-cut β -Ga₂O₃ surface.

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Current status of NanoTerasu BL06U: beamline for micro- and nano-ARPES measurements

M. Kitamura¹, F. Nishino¹, H. Iwasawa¹, N. Inami¹, T. Takeuchi¹, T. Imazono¹, Y. Ohtsubo¹,
J. Miyawaki¹, K. Yamamoto¹, K. Inaba¹, A. Agui¹, T. Nakatani¹, M. Tsukamoto¹,
K. Fujii¹, H. Kimura¹, and K. Horiba¹

¹ NanoTerasu Center, National Institutes for Quantum Science and Technology (QST)

Angle-resolved photoemission spectroscopy (ARPES) is a powerful experimental technique for directly probing the electronic band structure of materials and has been extensively utilized in the study of various oxide systems [1-3].

The beamline BL06U is one of the public beamlines at NanoTerasu, a 3-GeV highbrilliance synchrotron radiation facility at Tohoku, Japan. This beamline is designed for ARPES measurements using micro-focused and nano-focused beams, including spin-resolving. A 4 m long APPLE-II type undulator provides brilliant soft x-rays with various polarizations (linear horizontal and vertical, and left and right circular) in the 50–1,000 eV energy range. A collimated plane grating monochromator (cPGM) is employed as a monochromator [4]. We have been commissioning the beamline optics, and the energy-resolution $E/\Delta E$ has achieved the designed value (about 60,000 at 65 eV), which was evaluated by multiple ionization spectra of helium gas.

BL06U can be operated by switching between two branches. In the B-branch, a focused beam of less than 10 μm has been achieved using a Wolter-I mirror, and user operation as a micro-ARPES station began in March 2025. Meanwhile, the nano-ARPES station is under the construction in the A-branch. Unlike many other nano-ARPES stations that use Fresnel zone plates for nanoscale beam focusing, this nano-ARPES station adopts an ultra-high-precision mirror optical system. This approach aims to generate a high-flux nano-focused beam and achieve highly efficient ARPES measurements. As a first step, a 1 μm focused beam using a Wolter-I mirror will be realized and make available for user operation. In the next step, the goal is to achieve soft X-ray ARPES measurements using about a 100 nm focused beam with a developed tube-shaped mirror. In this presentation, we report on the current commissioning status of the BL06U beamline, with a primary focus on the progress of the A-branch nano-ARPES station.

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Growth condition optimization for various chromium-oxide films by using an in-situ MBE-ARPES system

Hanjun Sim¹ and Byungmin Sohn¹

¹*Department of Physics, Sungkyunkwan University, Suwon, Republic of Korea*

Chromium-based oxides show various intriguing phenomena such as half metallicity in CrO₂ [1], antiferromagnetism in Cr₂O₃ [2], and giant spin splitting in a CrO monolayer [3]. The phases of these compounds are very sensitive to the growth conditions, especially to the oxidation condition. Here, we investigate the growth conditions for various chromium-based oxides by using an *in-situ* molecular beam epitaxy (MBE)–angle-resolved photoemission spectroscopy (ARPES) system. Chromium-based thin films are grown in our ozone-assisted MBE system, and the electronic structures are characterized by in-situ ARPES. A reflection high energy electron diffraction (RHEED)-based analysis is also developed to support the optimization of the growth conditions.

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Resonant soft x-ray scattering on layered nickelates and oxide interface

Ronny Sutarto¹, Robert Green², David G. Hawthorn³, Teak Boyko¹

¹ *Canadian Light Source, Saskatoon, Saskatchewan S7N 2V3, Canada*

² *Department of Physics & Engineering Physics, University of Saskatchewan, Saskatoon, Saskatchewan S7N 5E2, Canada*

³ *Department of Physics & Astronomy, University of Waterloo, Waterloo, Ontario, N2L 3G1, Canada*

Layered nickelates have recently drawn much attention after superconductivity was discovered in infinite-layer of nickelate thin films. Furthermore, correlated electronic phases were also detected in other forms of bulk nickelates such as $\text{La}_3\text{Ni}_2\text{O}_7$ that would potentially indicate an analogy to cuprate superconductors. Both have layer structural patterns and nominal electronic configurations, however distinct magnetic and orbital physics are expected. Here we will showcase a few resonant soft x-ray scattering (RSXS) studies on layered nickelates and oxide interfaces at the Resonant Elastic and Inelastic X-ray Scattering (REIXS) beamline [1]. The beamline and endstation capabilities will be highlighted first.

For the first example, earlier reports indicated that a charge ordering phenomenon exists in an infinite-layer nickelate superconductor with its transition temperature to drop as a function of doping. By employing RSXS with an energy-resolved detector to separate out a substantial oxygen fluorescence background, it has been substantiated that the charge ordering is clearly absent in the pristine NdNiO_2 . Conversely, in the partially reduced NdNiO_2 , the ordering is instead linked to a structural modulation consisting of oxygen deficiencies. These findings clearly underline a contrasting phase diagram between copper and nickel-based superconductors [2].

Secondly, the existence of spin ordering at a wave vector of $(H=0.25, K=0.25)$ with its transition around 150 K in $\text{La}_3\text{Ni}_2\text{O}_7$ has been recently reported to accompany superconductivity under hydrostatic pressure. Here, by means of azimuthal dependent RSXS, we could determine the spin structure, namely the magnetic moments are lying within the NiO_2 plane and perpendicular to the wave vector, in a remarkable agreement with diagonal, bicollinear spin stripe model [3].

Lastly, electronic reconstructions at the complex oxide $\text{LaAlO}_3/\text{SrTiO}_3$ interface have been revisited using resonant x-ray reflectometry (RXR). Having overcome the extrinsic factor of oxygen vacancies, we elucidated the existence of the half charge at a confined reconstructed interface that is fully consistent with the theory of polar catastrophe [4].

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in situ Synchrotron-Radiation Photoemission Spectroscopy of NbO₂ Ultrathin Films

H. Konaka¹, D. Shiga^{1,2}, S. Inoue¹, Y. Masutake¹, Z. Zhang¹, T. Soma¹, K. Ozawa², and H. Kumigashira^{1,2}

¹ IMRAM, Tohoku University, Sendai, Japan

² Photon Factory, IMSS, KEK, Tsukuba, Japan

The transition-metal oxide NbO₂ with a $4d^1$ electronic configuration is in a body-centered tetragonal (BCT) insulating phase with Nb-Nb dimerization at room temperature. It undergoes a transition to a rutile-type metallic phase at approximately 1080 K. Recently, the formation of an energy gap associated with the structural phase transition was observed by photoemission spectroscopy [1]. Based on this observation and previous theoretical work [2], the BCT phase is understood as a Peierls insulating phase accompanied by Nb-Nb dimerization. Meanwhile, VO₂, which has a $3d^1$ electronic configuration, exhibits a similar Peierls transition accompanied by V-V dimerization; however, an electronic phase without V-V dimerization has been reported to emerge in the ultrathin-film limit, where the film thickness (t) is reduced to a few atomic layers [3]. Therefore, it is expected that reducing the dimensionality of NbO₂ thin films would destabilize Nb-Nb dimerization, leading to the emergence of a metallic electronic state. In this study, we fabricated thickness-controlled epitaxial NbO₂ ultrathin films and investigated their electronic structure on the few-nanometer scale using *in situ* synchrotron-radiation photoemission spectroscopy.

Figure 1 shows the thickness dependence of the *in situ* valence-band spectra of NbO₂ ultrathin films measured at 20 K. For the thick film ($t = 20$ nm), a peak structure originating from the Nb $4d$ states exists at approximately 1.4 eV, indicating the formation of a Peierls gap near the Fermi level (E_F), which is characteristic of the BCT insulating phase in NbO₂ [1]. Contrary to the expectation, even when the film thickness is reduced to the ultrathin-film regime, this peak position remains almost unchanged, and the emergence of spectral weight near E_F , indicative of metallization, is not observed. These results suggest that the Nb-Nb dimerization in NbO₂ is robustly maintained even in the ultrathin-film regime, in contrast to VO₂ [3].

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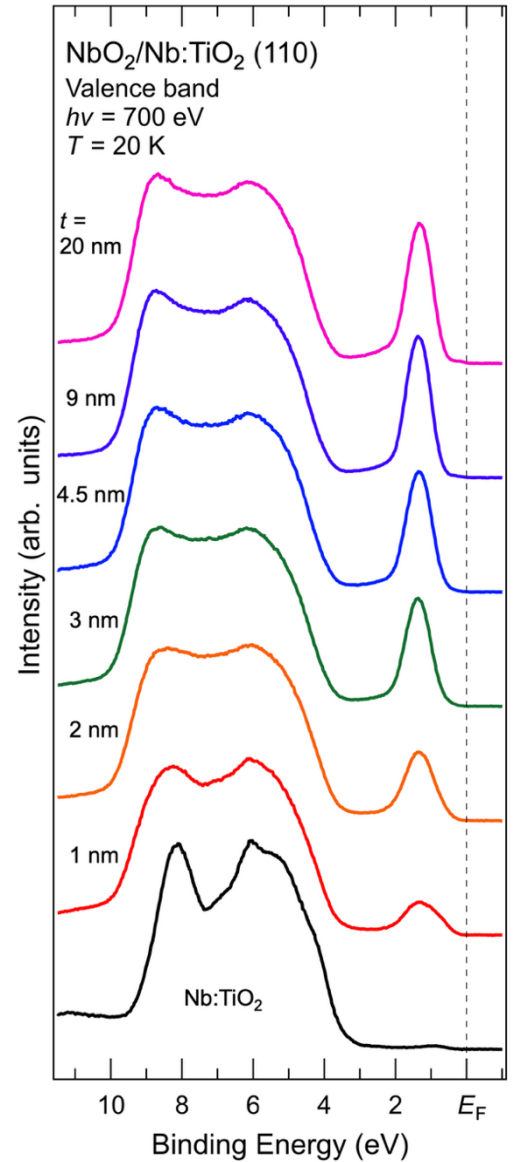


Figure 1: Thickness dependence of *in situ* valence-band spectra for NbO₂/Nb:TiO₂ (110) ultrathin films.

The Oxidation Behavior of Nb oxides seen by X-ray Photoemission Spectroscopy

Jasnamol Palakkal^{1,2}, Pia Henning¹, and Lambert Alff²

¹ *Institute of Materials Physics, Georg-August-University of Göttingen, Germany*

² *Institute of Materials Science, Technical University of Darmstadt, Germany*

Nb-based compounds are very important in many applications like superconductivity, energy storage and conversion devices, wireless communication systems, etc. [1-3]. Many of these functionalities are correlated to the electronic state of Nb. A general tool that materials scientists use to determine oxidation states is X-ray Photoemission Spectroscopy (XPS). When assuming the cationic state of the complex metallic/semiconducting Nb^{x+} using such techniques, final-state plasmonic effects need to be accounted for [4]. Recently, we investigated the off-stoichiometry engineering of transparent conducting SrNbO₃ for tuning its optical and electrical properties [5]. In these samples, the density of states (DOS) near the Fermi level led to effects similar to those of plasmonic final-state and to the appearance of an additional peak in the XPS at a lower binding energy than that of Nb⁴⁺ [5]. Such a peak was missing on the films deposited with a Pt capping layer [6]. Various studies in literature either attributed this peak to another oxidation state or to an artifact. In order to confirm, we have deposited a series of partially oxidized NbO_x samples using molecular beam epitaxy (MBE), and performed XPS [7].

By comparing the XPS spectra for the prepared NbO_x films with those for metallic and insulating Nb-containing perovskite oxides, we confirmed that final-state effects are present in these complexes when the DOS near the Fermi level is sufficiently occupied, thereby interacting with the photoion [7]. This observation is further substantiated by our recent work [unpublished], in which oxygen doping effects SrNbO_{3+δ} altered the metallic conductivity. In SrNbO_{3+δ}, the material becomes semiconducting upon oxygen introduction, with the DOS near the Fermi level increasing due to intrinsic defects. This, in principle, caused the plasmonic effects to appear more prominent in SrNbO_{3+δ}, even more than that of the metallic SrNbO₃. During the talk, I will summarize our results by comparing and reviewing different example spectra of Nb complexes from the literature.

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Exploring surface electronic structure in titanates by in situ synchrotron resonant x-ray reflectometry

Jessica Freese¹, Ronny Sutarto², Robert Green¹

¹ *University of Saskatchewan, Saskatoon, SK, Canada*

² *Canadian Light Source, Saskatoon, SK, Canada*

Two-dimensional electron gases (2DEGs) have been observed not only at $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces, but at bare SrTiO_3 surfaces [1-3]. Examining the orbital reconstruction in SrTiO_3 surfaces and related materials provides a window into the tunability of these systems.

Here, we investigate the impact of crystal structure, A-site cation chemistry, and surface treatment on the electronic structure of titanate surfaces. Resonant x-ray reflectometry (RXR) is a synchrotron enabled technique which provides a depth-resolved probe sensitive to both element and electronic structure. In the soft x-ray regime, RXR offers a particular sensitivity to the Ti 3d orbital energies. Here, we present a layer-by-layer examination of the surface electronic structure in pure SrTiO_3 , 0.7 wt% Nb doped SrTiO_3 , CaTiO_3 , and BaTiO_3 . We also compare surfaces exposed to the ambient environment to surfaces transferred under vacuum immediately after annealing.

We also describe a robust approach to analysis of RXR data which sheds light on correlations between fitted parameters, verifies fit convergence, and establishes that the identified fit results represent a global minimum.

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Depth Profiling of Electron States at Buried Interface

Deok-Yong Cho¹

¹ *Department of Physics, Jeonbuk National University, Republic of Korea*

We investigated the quasi-free electron states at atomic-layer-deposited $\text{Al}_2\text{O}_3/\text{TiO}_2$ and $\text{Al}_2\text{O}_3/\text{ZnO}$ thin films heterostructures using hard X-ray photoemission spectroscopy. Utilizing the near-total-reflection geometry, the interface electronic structure near the Fermi level was successfully sketched with depth resolution under a nm. The results of X-ray-incidence-angle-dependent analyses reveal the depth-resolved details of the defect states clarifying their origins and provide a conclusive evidence of the nature of the quasi-2-dimensional electron gas at the both interfaces.

This work is done under many collaborations mainly with Dr. Cheng-Tai Kuo (SLAC, USA), Dr. Jean-Pascal Rueff (Soleil, France), Dr. Masato Yoshimura (SPring-8) and Prof. Tae Joo Park's group at Hanyang University, Korea.

Scan, Shift, Lock: Switching and Control of Ferroic Domain Walls via STEM Probe Trajectories

Mariana Palos¹, Noah Shnitzer¹, Fintan Hardy¹, Shelly Conroy¹

¹ Imperial College London, London UK

High-energy focused electron probes in scanning transmission electron microscopy (STEM) not only enable atomic-resolution imaging, but can also drive local changes in ferroic order [2,3]. Here, we demonstrate that the trajectory of the STEM probe provides a deterministic and tunable control parameter for manipulating and stabilizing ferroic domain wall configurations.

We investigate domain wall dynamics in copper–chlorine boracite ($\text{Cu}_3\text{B}_7\text{O}_{13}\text{Cl}$) and cryogenic BaTiO_3 using designed scan strategies. Systematic variation of probe trajectories produces reproducible and trajectory-dependent domain evolution. Conventional raster scans induce cumulative domain wall translation governed by scan direction and polarization orientation, while inward spiral scans produce an initial displacement followed by stabilization into a locked configuration. When domain walls remain within the field of view, switching between scan patterns restores prior states, demonstrating reversible and cyclical control of domain configurations, analogous to reversible switching pathways reported in ferroic systems [4,5].

Using 4D-STEM diffraction, we directly track symmetry changes associated with domain switching. In boracite, the emergence of ghost diffraction disks indicates polarization rotation away from the $[001]$ axis, consistent with continuous reorientation mechanisms previously discussed in ferroics [6]. In contrast, BaTiO_3 exhibits significantly higher sensitivity to experimental perturbations, where sub-nanometer drift or minimal specimen tilt suppresses Friedel-pair symmetry breaking, highlighting the stringent stability requirements for probing switching pathways in oxide ferroelectrics.

These results establish electron probe trajectory design as a route for controlled manipulation of ferroic domain states, linking beam-driven dynamics with functional behavior. Beyond mitigating beam-induced artefacts, this approach enables the engineering of domain architectures and offers a pathway toward *in situ* control of ferroic functionality in oxide electronic materials.

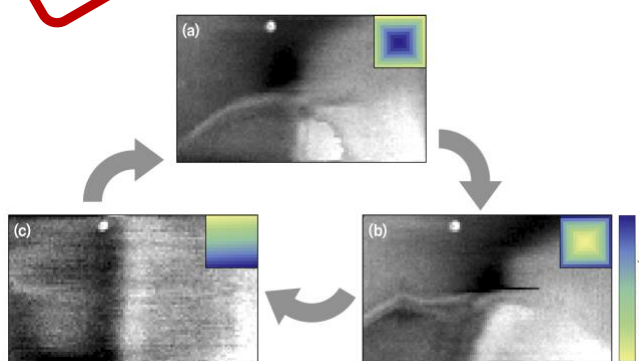


Figure 1: Domain reconfiguration controlled by STEM probe trajectory in $\text{Cu}_3\text{B}_7\text{O}_{13}\text{Cl}$ boracite. Virtual ADF images reconstructed from 4D-STEM datasets acquired using (a) inward spiral, (b) outward spiral, and (c) raster scans.

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Shubnikov-de Haas Oscillations Reaching the Quantum Limit in Two-Dimensional Electron Systems at SrTiO₃ (111) Interfaces

Ziqiao Wang¹, Autumn Heltman¹, Shalini Kumari¹, Lunhui Hu¹, Zhu Lin¹, Rojin Taheri², Leixin Miao², Nasim Alem², Lin Jiao³, Shalinee Chikara³, Alexey Suslov³, John Singleton⁴, Fedor Balakirev⁴, Chaoxing Liu¹, Qi Li¹

¹ *Department of Physics, The Pennsylvania State University*

² *Department of Materials Science and Engineering, The Pennsylvania State University*

³ *National High Magnetic Field Laboratory, Tallahassee*

⁴ *National High Magnetic Field Laboratory, Los Alamos National Laboratory*

Transition metal oxides oriented along the (111) direction have been predicted to host exotic topological phases and unconventional quantum states due to their unique hexagonal crystal symmetry and the interplay between charge, spin and orbital degrees of freedom. Despite these fascinating predictions, experimental probe of Fermi surface through transport measurements has remained challenging due to the difficulty in fabricating high-mobility samples. In this work, we report the observation of pronounced Shubnikov-de Haas (SdH) oscillations extending into the quantum limit at magnetic fields up to 35 T. These quantum oscillations were realized in high-mobility ($>20,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) two-dimensional electron liquids at (111)-oriented SrTiO₃ interfaces with tunable carrier densities. At low Landau levels, clear spin splitting is observed, which is attributed to the interplay between Zeeman splitting and Rashba spin-orbit coupling according to our theoretical model. This quantitative analysis yields a Landé g-factor of 0.29 and a Rashba coefficient $\alpha = 0.6 \text{ meV nm}$. Remarkably, upon entering the quantum limit where only the lowest Landau level is occupied, the transport behavior exhibits a dramatic transition. The magnetoresistance evolves into a giant, non-saturating linear field dependence, while the temperature dependence of the sheet resistance reveals a metallic to insulating state transition. These findings suggest that the strongly correlated electron system at the SrTiO₃ (111) interface could give rise to novel insulating states in the quantum limit.

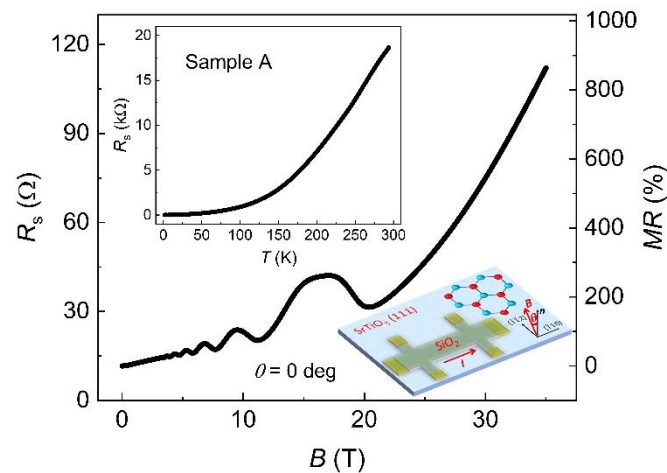


Figure 1: Pronounced SdH oscillations extending into the quantum limit at (111)-oriented SrTiO₃ interfaces [1].

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Lowest Proton Diffusion Barrier in EuNiO_3 Revealed by DFT and EIS

Kenta Nakanishi¹, Musa Alaydrus³, Ikutaro Hamada^{2,3}, Li Haobo^{1,2}, Hidekazu Tanaka^{1,2}

¹ *SANKEN (The Institute of Scientific and Industrial Research), University of Osaka*

² *Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives (OTRI), University of Osaka*

³ *Department of Precision Engineering, Graduate School of Engineering, University of Osaka*

E-mail: kenta_nakanishi77@sanken.osaka-u.ac.jp

Perovskite rare-earth nickelates, such as SmNiO_3 , are known as Mott-transition materials whose electronic states can be controlled by proton insertion [1]. This characteristic makes them promising candidates for iontronic devices such as resistive random-access memory (ReRAM) [2]. However, because ionic devices generally operate more slowly than electronic devices, the development of materials with faster proton transport and improved switching performance is highly desirable.

In this study, proton diffusion in RENiO_3 (RE = Nd, Sm, and Eu) was investigated using both density functional theory (DFT) calculations and electrochemical impedance spectroscopy (EIS). Figure (a) shows the proton migration pathway in RENiO_3 , and Fig. (b) presents the corresponding relative potential energy profiles calculated by the nudged elastic band method. The calculations reveal that inter-octahedral hopping along the [110] direction is the dominant diffusion bottleneck and that EuNiO_3 exhibits the lowest proton migration barrier among the investigated RENiO_3 compounds.

To experimentally verify the theoretical prediction, EIS measurements were performed on EuNiO_3 thin films grown on LaAlO_3 , LSAT, and SrTiO_3 substrates. As shown in Fig. (c), the activation energy for proton diffusion was estimated to be 0.201 eV for $\text{EuNiO}_3/\text{LaAlO}_3$, which is significantly lower than those obtained for the other substrates. Figure (d) summarizes the dependence of proton diffusion activation energy on the rare-earth ionic radius. The reduction in activation energy correlates with the decrease in rare-earth ionic radius. Both the DFT calculations and experimental results consistently indicate that EuNiO_3 possesses the lowest proton diffusion barrier among the RENiO_3 family.

These findings demonstrate that EuNiO_3 is a promising material for high-speed iontronic devices and provide a design guideline for fast-switching proton-conducting oxides.

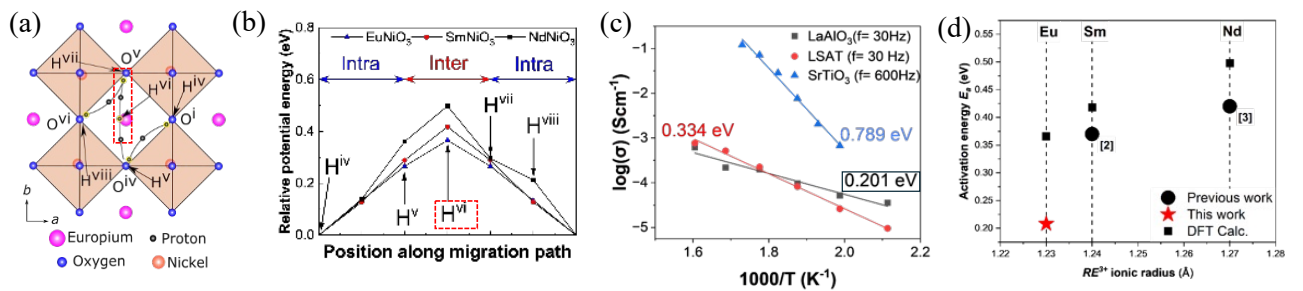


Figure (a) The schematic diagram of proton migration path in RENiO_3 and relative potential energy of proton, (b) RE-ionic radii dependence of activation energy of proton diffusion, (c) Arrhenius plot for EuNiO_3 thin films on LaAlO_3 , LSAT and SrTiO_3 substrates respectively, (d) RE-ionic radii dependence of activation energy of proton diffusion.

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In-situ Strain Engineering of Complex Oxide Membranes

Merit Spring¹, Varun Harbola², Yu-Jung Wu², Goro Nozue¹, Tien-Lin Lee³, Jochen Mannhart²,
Michael Sing¹, Ralph Claessen¹

¹ *Physikalisches Institut and Würzburg-Dresden Cluster of Excellence ctd.qmat, 97074 Würzburg, Germany*

² *Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany*

³ *Diamond Light Source Ltd., Harwell Science and Innovation Campus, Didcot, Oxfordshire OX11 0DE, UK*

Freestanding oxide membranes have recently emerged as a versatile platform for the design and control of novel electronic phases. Their fabrication relies on a sacrificial layer (SL), such as $\text{Sr}_3\text{Al}_2\text{O}_6$, which is epitaxially grown on a substrate. A thin film, destined to become the membrane, is then epitaxially deposited on top of this SL. Owing to the solubility of the SL – $\text{Sr}_3\text{Al}_2\text{O}_6$, for example, dissolves in water – it can be selectively removed, allowing the membrane to be released and subsequently transferred onto a different platform.

The concept of membranes provides access to interfaces with broken symmetries that cannot be realized by conventional epitaxial growth [1,2], enables substrate-free characterization techniques [3] and allows virtually any material to serve as carrier for a vector substrate, i.e., a membrane, which is chemically and crystallographically independent of the carrier's bulk [4]. Beyond these remarkable opportunities, the flexibility of oxide membranes makes them particularly well suited for the application of external strain [5].

In this work, strain is applied to oxide membranes to tailor their electronic properties and potentially drive Mott type metal-insulator transitions in d^1 transition-metal oxides such as SrVO_3 (SVO) and LaTiO_3 (LTO). To achieve this, the membranes are transferred directly onto an in-situ strain device developed at beamline I05 of the Diamond Light Source. The application of strain is verified through synchrotron-based X-ray Bragg measurements, which provide direct access to changes in the lattice parameters. The resulting modifications of the electronic structure are then probed by photoemission spectroscopy (PES). In initial measurements on SrTiO_3 membranes, we observe a shift of the Ti 2p core level to higher binding energies under tensile strain (Fig. 1), reflecting a modified balance between ionicity and lattice potential at the Ti sites, driven by variations in bond lengths, and structural changes, respectively.

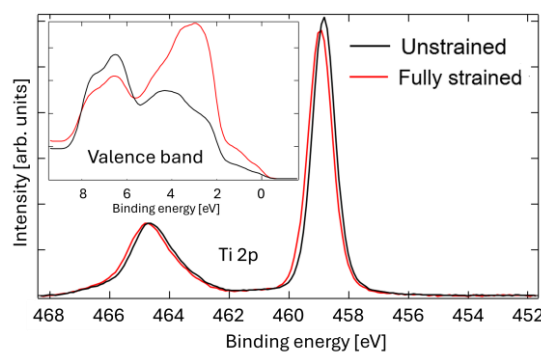


Figure 1: Strain-dependent Ti 2p PES spectra of a 120nm thick STO membrane.

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Quantum dots in Freestanding $\text{LaAlO}_3/\text{SrTiO}_3$ Micro-membranes

G. Meucci¹, P. O. Konstantinopoulou¹, T. Jansen¹, G. Nagda¹, D. J. Carrad¹, E. Di Gennaro², Y. Chen³, N. Manca³, N. Bergeal⁴, M. Bibes⁵, A. Kalaboukhov⁶, M. Salluzzo³, R. Citro³, F. Trier¹, N. Pryds¹, F. Miletto Granozio³, A. Sambri³, and T. S. Jespersen¹

¹ Technical University of Denmark, 2800 Kgs. Lyngby, Denmark

² Università degli Studi di Napoli “Federico II”, 80126 Naples, Italy

³ CNR-SPIN, 80126 Naples and 16152 Genova and 84084 Fisciano, Italy

⁴ ESPCI Paris, Université PSL, CNRS, Sorbonne Université, Paris, France

⁵ Université Paris-Saclay, 91767 Palaiseau, France

⁶ Chalmers University of Technology, 41296 Gothenburg, Sweden

Freestanding oxide membranes are attracting significant interest for integrating complex oxides with semiconductors and, when scaled to the micron scale, provide a platform for quantum devices. In particular, freestanding $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) micro-membranes (Fig. 1a) enable the study of correlated electron phenomena, including unconventional electron pairing without superconductivity, which in confined systems is described by a negative effective charging energy (negative- U).

We investigate negative- U quantum dots in freestanding LAO/STO micro-membranes on Si/SiO_2 (Fig. 1b). We find that these membranes naturally host negative- U centers (Fig. 1c,d), where confinement arises intrinsically, rather than from engineered nanostructures, coexisting with gate-tunable superconductivity and stable under thermal cycling. Our results suggest that negative- U centers arise from intrinsic interfacial inhomogeneity and may be a general feature of LAO/STO [1].

We compare the intrinsic confinement centers with depletion- and gate-defined quantum dots. Notably, we demonstrate the first LAO/STO quantum dot devices on silicon, highlighting the potential of freestanding membranes for oxide-semiconductor integration.

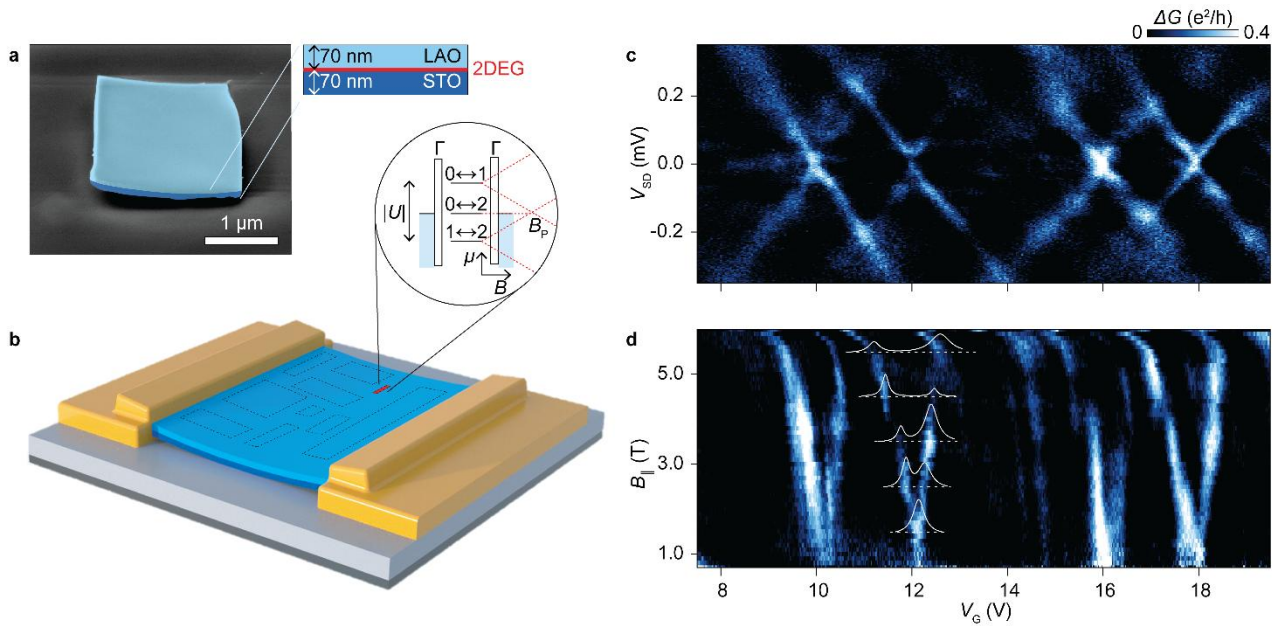


Figure 1: a Scanning electron micrograph of a membrane on SiO_2 . b Device schematic with a representation of intrinsic LAO/STO inhomogeneity. c Residual conductance, ΔG , vs gate voltage V_G and bias voltage V_{SD} . d ΔG vs V_G and $B_{||}$.

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Local Strain Engineering of Freestanding Crystalline SrTiO₃ Membranes

Laszlo Cline¹, Jennifer Fowlie¹

¹ Northwestern University Department of Materials Science and Engineering

Transition metal oxides frequently exhibit strongly correlated lattice, electronic, and magnetic degrees of freedom. The standard approach to investigating these correlations in thin films—substrate-imposed epitaxial strain—is limited by the discrete lattice parameters of available substrates. Recent advances in sacrificial layer-enabled exfoliation of freestanding crystalline oxide membranes [1] overcome this issue, providing access to a wide, continuous range of strain states. While most oxide membrane strain engineering studies involve the application of global uniaxial or biaxial strain [2, 3], we instead leverage lithography and etching techniques to locally engineer the surface texture of the Si host substrate, imposing unique, customizable mechanical boundary conditions [Figure 1A, 1B]. SrTiO₃ is an ideal model system for such investigations; it is a quantum paraelectric with incipient ferroelectric order suppressed by its centrosymmetric cubic perovskite structure. However, this symmetry constraint can be lifted at room temperature with just 1-2% tensile strain [3, 4]. We transfer SrTiO₃ membranes onto textured Si and predict resultant in-plane bending strains of up to 2.33% using finite element analysis [Figure 1C]. This substrate texture-imposed strain is likely sufficient to break inversion symmetry and locally stabilize ferroelectricity, as observed by Xu *et al.* in globally strained SrTiO₃ membranes with piezoresponse force microscopy and second harmonic generation [4]. We will present our efforts to elucidate the influence of heterogeneous, substrate-imposed strain on SrTiO₃ membrane structure and properties.

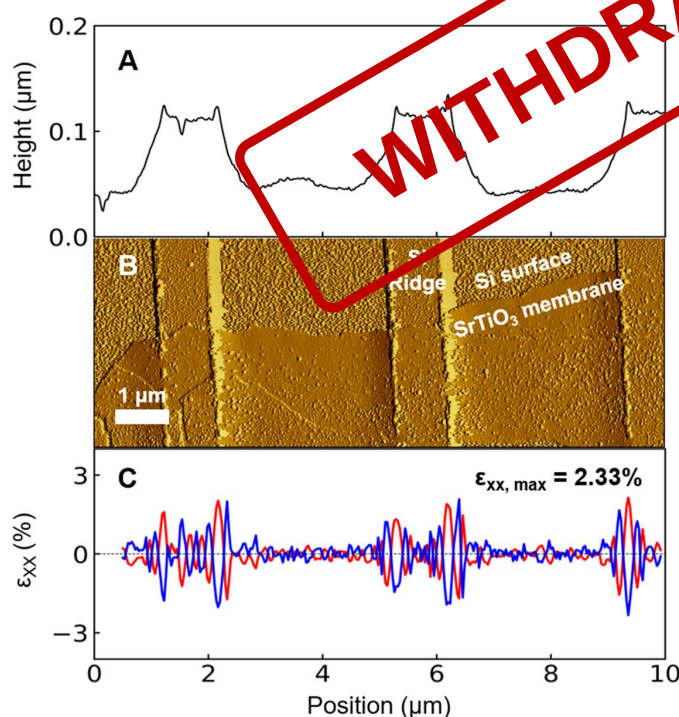


Figure 1. A: Atomic force microscopy (AFM) line profile of a SrTiO₃ membrane (~13 nm thick) strained by Si host substrate ridges. B: AFM amplitude error image of the strained membrane. C: Predicted membrane strain profile from finite element analysis of the line profile in A.

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Structural phase transformation freestanding zirconia membranes

S. Shindo¹, K. Miyazaki¹, R. Maki², M. Nakahata³, O. Ikeda⁴, T. Koganezawa⁴,
K. Okamoto³, H. Funakubo³, Y. Shimakawa², D. Kan¹
Univ of Osaka¹, Kyoto Univ.², Science Tokyo.³, JASRI⁴

Freestanding and crystalline membranes fabricated by exfoliating epitaxial films from substrates provide a platform for creating heterostructures unattainable by conventional thin-film growth techniques. We found that metastable phases of ZrO_2 (ZO) can be maintained even after exfoliation from the substrate, and that the membranes exhibit a relatively high dielectric constant of approximately 20 [1]. However, the factors that maintain the metastable structure remain elusive. In this study, we used synchrotron X-ray diffraction to clarify how the exfoliation process affects the structural properties of ZO membranes.

ZO membranes were prepared from ZO epitaxial films grown on SrTiO_3 (STO) substrates. The ZO epitaxial films were deposited by pulsed laser deposition on (100) and (110) STO substrates buffered with $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) sacrificial layers and were then released from the substrates by selectively etching the LSMO layers. Figure 1 shows synchrotron films and the freestanding membranes. The using 12.4 keV X-rays. The diffraction im collected while rotating the sample throug on the (100) STO substrate, which consist the diffraction images before and after metastable phases were maintained even in substrate, which consists of the metastable spots from the (111) metastable phase be addition, reflections from the monoclinic of ZO, were observed, indicating the Furthermore, our results suggest that grain and (001)-oriented metastable phases ma structures of ZO membrane.

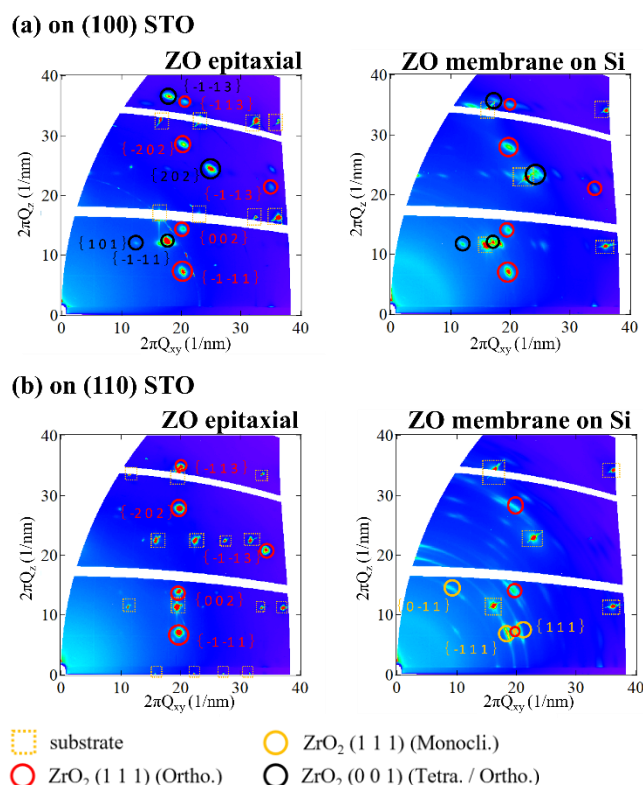


Figure 1: Synchrotron X-ray diffraction images for 20 nm-thick ZrO_2 epitaxial films on (100) and (110) STO substrates and ZO membranes fabricated by exfoliating them from STO substrates and transferring them on Si substrates.

LSMO layers. Figure 1 shows synchrotron X-ray diffraction images of 20-nm-thick ZO epitaxial films and the freestanding membranes. The measurements were performed at BL13XU of SPring-8 using 12.4 keV X-rays. The diffraction images were obtained by integrating the diffraction patterns collected while rotating the sample through 360° around the ϕ axis. For the ZO epitaxial film grown on the (100) STO substrate, which consist of metastable ZO phases with (111) and (001) orientations, the diffraction images before and after exfoliation were almost identical, indicating that the metastable phases were maintained even in the membrane. In contrast, for the film grown on the (110) substrate, which consists of the metastable ZO phases with the (111) orientation only, the diffraction spots from the (111) metastable phase became ring-shaped diffraction patterns after exfoliation. In addition, reflections from the monoclinic structure, which is the thermodynamically stable structure of ZO, were observed, indicating the occurrence of structural relaxation in ZO membranes. Furthermore, our results suggest that grain boundaries resulting from the coexistence of the (111)- and (001)-oriented metastable phases may play an important role in maintaining the metastable structures of ZO membrane.

Solid-state electrochemical functionalization of YBCO free-standing membranes

MMBotana^{1,2}, J. Corral-Sertal^{1,2}, V. Álvarez-Martínez^{1,2}, V. Leborán^{1,2}, Rafael Ramos^{1,2}, F. Rivadulla^{1,2}

¹ Centro Singular de Investigación en Química Biolóxica e Materiais Moleculares (CIQUS),
Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain

² Departamento de Química-Física, Universidade de Santiago de Compostela, 15782 Santiago de Compostela, Spain

The integration of high-T_c cuprate superconductors in functional hybrid devices is especially constrained by the requirement of epitaxial growth and functionalizing post-treatments of the individual constituents. An effective way to deal with this issue is to synthesize freestanding membranes of these oxides, which allows the use of lattice-matching substrates, yielding high-quality thin films. On the other hand, in cuprates like YBaCuO, the superconducting properties are determined by the oxygen content. In this work, we develop a methodology for tuning the oxygen concentration of freestanding membranes via electrochemical redox reactions. We first optimized the growth and release of large-area YBCO freestanding membranes of high crystalline quality [1]. The membranes are then transferred onto a solid-state electrolyte substrate, YSZ, which allows the electric-field driven control of the oxygen content of the YBCO membranes [2]. In situ X-ray diffraction and electrical conductivity demonstrate the reversibility of the process, opening new possibilities in the fabrication of complex heterostructure devices based on high-T_c superconductors.

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Polar Textures from Ferroelastic Topology in a Metal Membrane

Rahil Haria¹, Noah Schnitzer¹, T. Ben Britton^{1,2}, Yaqi Li¹, Tom J. P. Irons³, Sophia Linssen-Pitsaros^{1,4,5}, Ella Banyas^{6,7}, Geri Topore¹, Annabel Hoyes¹, Mariana Palos¹, Sinéad M. Griffin^{6,7}, Katherine Inzani³, and Michele Shelly Conroy¹

¹ *Department of Materials, Imperial College London, London, SW7 2AZ, United Kingdom*

² *Department of Materials Engineering, UBC, Vancouver, British Columbia, Canada*

³ *School of Chemistry, University of Nottingham, Nottingham NG7 2RD, UK*

⁴ *Department of Physics and Astronomy, University College London, London WC1E 6BT, United Kingdom*

⁵ *London Centre for Nanotechnology, 17–19 Gordon Street, London WC1H 0AH, United Kingdom*

⁶ *Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

⁷ *Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

Polar metals, which combine electric polarisation with metallic conduction, are exceptionally rare because itinerant electrons screen long-range dipoles and favour centrosymmetric structures. In a ferromagnetic metal, where polarisation must also coexist with magnetic order, it is rarer still, and has previously required chemical symmetry breaking or engineered interfaces that localise the response [10]. Here we demonstrate that ferroelastic domain topology alone generates pervasive, intrinsic polar textures throughout a single freestanding membrane of the ferromagnetic metal SrRuO₃ [1,2], without chemical or interfacial symmetry breaking.

Using correlative ECCI and atomic-resolution STEM with Fourier-space strain analysis and DFT, we resolve two distinct origins of polarisation in a dense ferroelastic nanodomain network [5]. Polarisation emerges only at translation-inequivalent (hard) antiphase boundaries, not at translation-equivalent (easy) ones. This is governed by the multicomponent $a^-a^+c^+$ tilt order parameter: Néel-like interpolation at hard boundaries preserves the in-phase M_3^+ component where tilt gradients are steepest, amplifying roto-flexoelectric coupling [6] and producing displacements exceeding 13 pm, whereas Ising-like collapse at easy boundaries suppresses it. The polar character of a wall is therefore a boundary property, set by the domain translation rather than the host material, and is directly addressable by domain engineering. A second mechanism operates at embedded 90° ferroelastic walls, which nucleate ~4 nm polar nanoclusters via strain gradients from eigenstrain mismatch and rotostriction, forming divergent and convergent configurations resembling topological polar textures [8,9].

These results establish ferroelastic domain topology as a general, geometry-driven route to polar order in a ferromagnetic metal, replacing the chemical asymmetry and interfaces previously required, and extend the polar antiphase-boundary framework from insulating antiferroelectrics [7] to metals. Because the response is gated by domain symmetry and driven by local strain gradients, it is intrinsically tunable, coupling polarisation to strain and magnetism and offering a prospective route to reconfigurable Berry-curvature and spintronic control [3,4].

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Guidelines for sputtering growth of strontium aluminate and ferromagnetic $\text{Sm}_2\text{NiMnO}_6$ membranes

Evgenios Stylianidis, Marta Gibert

Institute of Solid State Physics, TU Wien, Vienna 1040, Austria

The recent advances on the research of free-standing transition metal oxides is largely owned to the development of perovskite-compatible materials which can be grown epitaxially as sacrificial layers and dissolved selectively in common solvents such as water. Among the various sacrificial materials developed to date, cubic $\text{Sr}_3\text{Al}_2\text{O}_6$ [1] is one of the most notable examples. The alternative lower symmetry tetragonal $\text{Sr}_4\text{Al}_2\text{O}_7$ phase has shown enhanced structural flexibility [2,3].

After a few years of extensive studies, the most reported techniques for the growth of high-quality sacrificial layers are pulsed laser deposition and molecular beam epitaxy. In this work, we present the growth of strontium aluminate thin films via RHEED-assisted off-axis RF magnetron sputtering. To our knowledge, the group of Xia Hong has previously reported the growth of $\text{Sr}_3\text{Al}_2\text{O}_6$ with sputtering [4,5], however, a systematic study of the growth dependence on the deposition conditions is still lacking. Here, we investigate how the temperature, the partial pressure and the Ar/O_2 ratio influence the growth and crystallization of strontium aluminate. We demonstrate that the cubic $\text{Sr}_3\text{Al}_2\text{O}_6$ and the tetragonal $\text{Sr}_4\text{Al}_2\text{O}_7$ phases can be selectively stabilized by tuning the growth conditions (see Fig. 1), while using the same sputtering target, providing versatility over the lattice parameter mismatch.

Using our developed growth protocol we then released and transferred membranes of the double perovskite $\text{Sm}_2\text{NiMnO}_6$ onto support substrates. We show that the transferred films display good structural quality and rock-salt ordering of the Ni^{2+} - Mn^{4+} cations and retain the bulk ferromagnetic transition temperature.

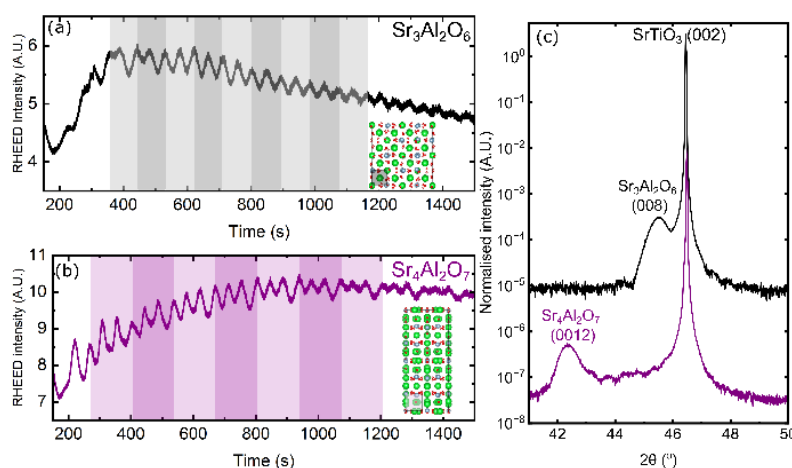


Figure 1: RHEED intensity as a function of time during the sputter growth of (a) cubic $\text{Sr}_3\text{Al}_2\text{O}_6$ and (b) tetragonal $\text{Sr}_4\text{Al}_2\text{O}_7$ films. (c) XRD of the respective films.

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Transfer of ZnO Thin Films Enabled by a Two-Step Sacrificial Layer Approach for Functional Hexagonal Crystal Films

Hiroaki Nishikawa¹, Yuya Oda²

¹ Faculty of Biology-Oriented Science and Technology, Kindai University

² Graduate School of Biology-Oriented Science and Technology, Kindai University

$\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) is an oxide that readily dissolves in pure water [1], and it has been widely used as a sacrificial layer for transferring functional oxide thin films onto other substrates. However, most of the functional oxides transferred using SAO are perovskite-type materials, which exhibit good lattice matching and structural compatibility with SAO. In contrast, we have initiated new research aimed at developing a transfer technique for hexagonal materials.

ZnO is a hexagonal oxide that exhibits c-axis preferred orientation regardless of the underlying substrate and is known to dissolve in a mixed solvent consisting of 60 vol.% methanol and 40 vol.% acetylacetone (acac) [2]. Taking advantage of this solubility, we propose a transfer method for functional hexagonal oxide thin films using an SAO/ZnO sacrificial layer structure. Specifically, SAO is deposited on a substrate (Fig. a), followed by ZnO (Fig. b), and then a functional hexagonal oxide is deposited on top (Fig. c). A polymer sheet is subsequently attached to the top surface (Fig. d). Immersing the fabricated multilayer thin film in pure water dissolves the SAO layer (Fig. e), allowing the functional hexagonal oxide/ZnO structure to be separated from the substrate. The detached thin film is then immersed in acac to dissolve the ZnO layer (Fig. f), resulting in a flexible functional hexagonal oxide thin film with its surface exposed (Fig. g).

A key to this transfer process is the deposition of epitaxial ZnO (001) on the SAO layer. Therefore, in this study, we report on the fabrication and transfer of (001)-oriented ZnO thin films grown on perovskite (111) substrates with SAO (111) buffer layers.

SrTiO_3 (STO) (111) substrates were used as the perovskite-type substrate, on which (111)-oriented SAO was deposited by pulsed laser deposition, followed by (001)-oriented ZnO deposition. A polyethylene (PE) sheet was then attached, and the sample was immersed in pure water to dissolve the SAO layer, resulting in the separation of the STO substrate from the PE/ZnO structure. We report the results of X-ray diffraction $2\theta/\theta$ measurements on the fabricated ZnO/SAO/STO sample.

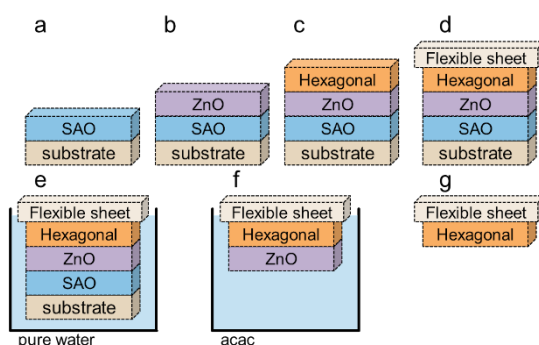


Figure Schematic diagram of the transfer process proposed for fabricating flexible thin films of hexagonal oxides.

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Highly Conductive Single-Crystalline Nb-Doped TiO₂ Membranes and Photochemically Accelerated Lift-Off for Transparent Oxides

Yuzuki Uchida¹, Jiyang Huang¹, Ryo Ishizeki¹, Satoshi Sasaki¹, Masamichi Negishi¹,
Seung Sae Hong², Tomoteru Fukumura^{1,3,4}

¹ Department of Chemistry, Graduate School of Science, Tohoku University, Sendai, 980-8578, Japan

² Department of Materials Science and Engineering, University of California, Davis, California, 95616

³ WPI Advanced Institute for Materials Research, Tohoku University, Sendai, 980-8577, Japan

⁴ Center for Science and Innovation in Spintronics, Tohoku University, Sendai, 980-8577, Japan

Single-crystalline oxide membranes have attracted increasing attention because epitaxial oxide layers can be lifted off and transferred onto various substrates [1]. Transparent conducting oxide membranes are especially promising for flexible transparent devices and biosensors, but material choices remain limited [2]. Anatase Nb-doped TiO₂ (TNO) is a strong candidate owing to its high transparency, high conductivity comparable to Sn-doped In₂O₃, earth-abundant composition, chemical stability, and biocompatibility [3]. However, the commonly used Sr₃Al₂O₆ sacrificial layer is poorly lattice-matched with TNO, leading to cracks and degraded conductivity after release [4]. In this study, we employed La_{0.7}Sr_{0.3}MnO₃ (LSMO) as a lattice-matched sacrificial layer to suppress cracking and fabricate highly conductive TNO single-crystalline membranes [5].

TNO/LSMO heteroepitaxial thin films were grown on SrTiO₃(100) substrates by pulsed laser deposition. Subsequently, the TNO membranes were released by selectively dissolving the LSMO sacrificial layer in hydrochloric acid containing potassium iodide as a reducing agent and transferred onto sapphire or PET substrates. The transferred membranes exhibited low electrical resistivity comparable to that of epitaxial TNO thin films (Fig. 1), indicating that the lattice-matched LSMO sacrificial layer effectively suppresses crack formation and preserves the high conductivity of TNO. We also developed a photochemically assisted lift-off process using the photocatalytic activity of the SrTiO₃ substrate. Under UV irradiation, LSMO was dissolved in dilute hydrochloric acid without potassium iodide. This method achieved either a more than tenfold increase in dissolution rate or dissolution in an acid solution more than 100 times more dilute than the conventional process, while maintaining the high crystallinity of the membrane (Fig. 2).

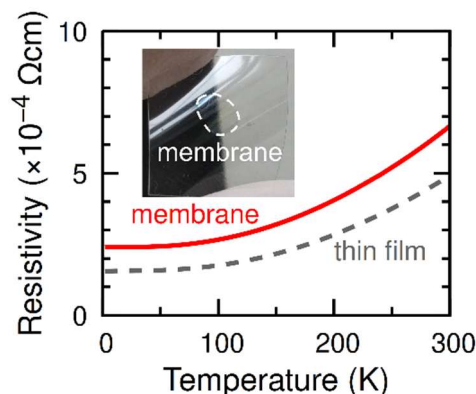


Figure 1: Electrical resistivity of the TNO epitaxial thin film and transferred membrane. The inset shows a photograph of the transferred membrane on a PET substrate.

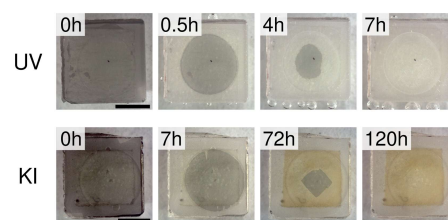


Figure 2: Time-dependent optical images showing the dissolution of the LSMO sacrificial layer by the photochemically assisted process (UV) and the conventional KI-based process (KI). Scale bar is 2 mm.

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van der Waals epitaxy of Ti_2O_3 films on hexagonal boron nitride toward freestanding oxide membranes

Keitaro Kanno¹, Takuto Soma², Satoru Fukamachi³, Hiroki Ago³, Daisuke Okuyama⁴,

Akira Ohtomo¹, Kohei Yoshimatsu¹

¹ Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan

² Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, Sendai, Japan

³ Faculty of Engineering Sciences, Kyushu University, Fukuoka, Japan

⁴ Institute of Materials Structure Science, KEK, Tsukuba, Japan

Corundum-type Ti_2O_3 exhibits a metal-insulator transition (MIT) at ~ 450 K, which is driven by lattice-deformation-induced $\text{Ti-}3d$ -band overlap [1]. Although this strong coupling between the structural and electronic properties is attractive for flexible electronics, Ti_2O_3 films grown on $\alpha\text{-Al}_2\text{O}_3$ (0001) substrates exhibit a lower MIT temperature (T_{MIT}) even below room temperature (RT), accompanied by an elongated c -axis due to compressive strain. The lowered T_{MIT} and non-flexible substrate hinder the applicability of Ti_2O_3 to flexible devices operable at RT. van der Waals (vdW) epitaxy using two-dimensional materials, which enables both strain relaxation and film exfoliation, is useful for overcoming these issues [2]. In this study, we grew Ti_2O_3 films on large-area hexagonal boron nitride (hBN) layers transferred to SiO_2/Si substrates using pulsed-laser deposition.

The reciprocal space map around the Ti_2O_3 $11\bar{2}6$ reflection (Fig. 1 (a)) indicated a distinct diffraction spot, confirming the epitaxial growth on hBN. A comparison of the lattice constants for various Ti_2O_3 (inset of Fig. 1 (a)) revealed that both the a - and c -axis lengths of the Ti_2O_3 films on hBN were closer to those of the bulk rather than those of $\text{Ti}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$ (0001). Reflecting the bulk-like lattice constants, the Ti_2O_3 films on hBN exhibited a T_{MIT} as high as 336 K, which is well above RT (Fig. 1 (b)). These results demonstrate that vdW epitaxy enables the nearly strain-free growth of Ti_2O_3 films, paving the way for flexible oxide electronics.

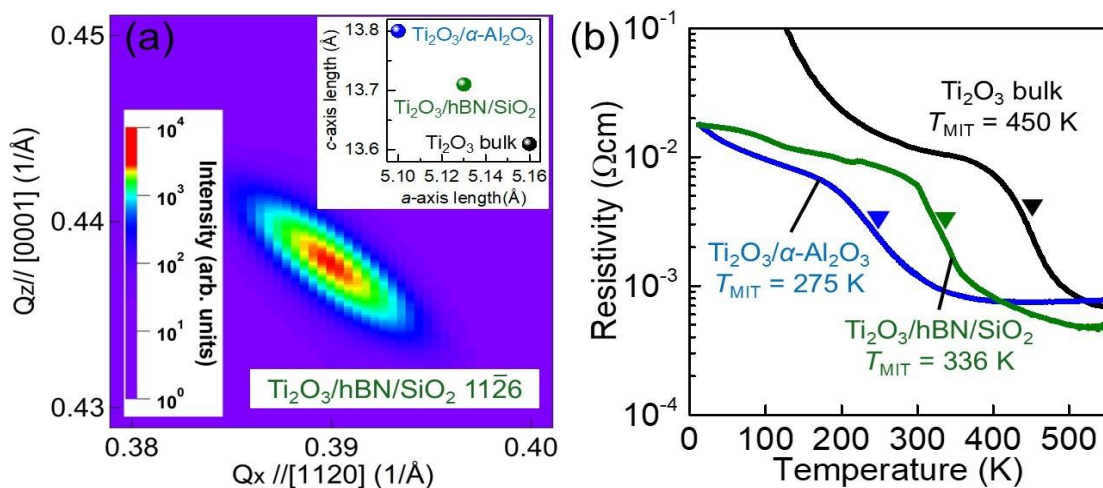


Figure 1. (a) Reciprocal space map around the $11\bar{2}6$ reflection of a Ti_2O_3 film grown on hBN/SiO_2 . The inset shows the lattice constants of various Ti_2O_3 samples at RT. (b) Temperature dependence of resistivity for $\text{Ti}_2\text{O}_3/\text{hBN}/\text{SiO}_2$, $\text{Ti}_2\text{O}_3/\alpha\text{-Al}_2\text{O}_3$, and bulk Ti_2O_3 [3]. The closed triangles indicate T_{MIT} .

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Transfer Approaches for Free-Standing Perovskite Oxide Membranes

Nicole Engel¹, Florian Hofmann¹, Benjamin Kunkel¹, Philipp Komissinsky¹, Juliette Cardoletti¹,
and Lambert Alff¹

¹*Institute of Materials Science, Technische Universität Darmstadt, Darmstadt, Germany*

The recent developments in membrane technology have the potential to upend the field of oxide electronics, opening new possibilities to create complex devices from oxides. In comparison with van der Waals-bonded materials, oxides display a wider range of functionalities and allow transfer of membranes with controlled thickness from the unit cell-level to hundreds of nanometers as well as of heterostructures. The challenge of combining highly crystalline functional oxide layers with other materials which cannot be grown in a conventional deposition process can be overcome using the wafer transfer technology.

Piezoelectric and ferroelectric membranes, in particular, have a strong potential for applications as their thin film counterparts have found their way into actuators, sensors, random access memories, etc. Literature reports exciting novel phenomena in ferroelectric thin films such as self-organized ripples. Yet, the effect of strain release in ferroelectric membranes and its impact on polarization and piezoelectric response remains under exploration.

Due to its highly tunable permittivity and low dielectric loss, lead-free barium strontium titanate (Ba,Sr)TiO₃ is widely used as a voltage-tunable capacitor (varactor) in various applications, including modulators, phase shifters, and waveguides. The ability to tune its Curie temperature through compositional changes of the Ba/Sr ratio allows also to tune its piezoelectric properties for a selected application.

In this work, we investigate the wafer transfer of (Ba,Sr)TiO₃ ferroelectric membranes. The (Ba,Sr)TiO₃ thin films, deposited by pulsed laser deposition on top of a Sr₂Al₃O₆ water-soluble sacrificial layer, are lifted-off from their GdScO₃ growth substrates. Using polydimethylsiloxane (PDMS) as a carrier wafer, the obtained membranes are transferred onto conductive substrates to enable electrical characterization, along with structural characterization by X-ray diffraction in order to compare the ferro-/piezoelectric properties of as grown thin films and their transferred membranes.

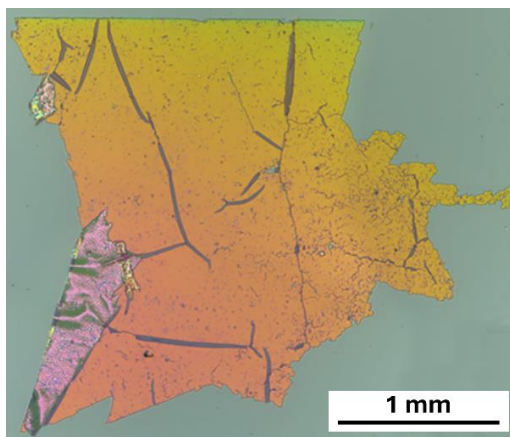


Figure 1: 100 nm-thick SrTiO₃ membrane transferred onto a Si wafer.

Internal Bias Relaxation and Defect-Mediated Polarization Instability in Ferroelectric $\text{Al}_{0.72}\text{Sc}_{0.28}\text{N}$ Thin Films

Seung Hyup Lee¹, Tae Yoon Lee¹, Jung Woo Cho¹, Chihwan An¹, and Seung Chul Chae^{1,2}

¹ *Department of Physics Education, Seoul National University, Seoul, 08826, Republic of Korea*

² *Center for Education Research, Seoul National University, Seoul, 08826, Republic of Korea*

Wurtzite-type $\text{Al}_{1-x}\text{Sc}_x\text{N}$ has attracted significant attention as a next-generation ferroelectric material due to its high remanent polarization and CMOS-compatible growth. The physical origins of field-cycling-induced wake-up and fatigue, however, remain insufficiently understood, limiting the reliable operation of $\text{Al}_{1-x}\text{Sc}_x\text{N}$ ferroelectric devices. In this work, we systematically investigate the evolution of domain dynamics and interfacial changes in $\text{Al}_{0.72}\text{Sc}_{0.28}\text{N}$ films during wake-up and fatigue behaviors. The films were deposited on Pt(111)/Ti/SiO₂/Si substrates by radio-frequency reactive sputtering at 300 °C. By combining first-order reversal curve, Rayleigh analysis, impedance spectroscopy, and X-ray photoelectron spectroscopy, we illustrated that wake-up and fatigue originate from distinct but competing processes during electric field cycling. During the wake-up stage, the formation of opposite-polarity nuclei against the as-grown preferred polarity increases the fraction of the film participating in polarization reversal, thereby enhancing switchable polarization. At higher cycle numbers, oxygen redistribution from the interface into the film lowers the effective dielectric response and is consistent with the formation of oxygen-incorporated regions with the degradation of ferroelectric properties. These regions suppress the field-dependent domain-wall response and make polarization switching more inhomogeneous, leading to fatigue behavior. These findings highlight the importance of interface and defect engineering for improving the reliability of nitride-based ferroelectric devices.

Boosting Inversion Symmetry Breaking in Epitaxial Tetragonal ZrO₂ via Atomic Layer Deposition

Jung Woo Cho¹, Dongmin Kim^{2,3,4}, In Hyeok Choi⁵, R. A. Madusanka Pushpakumara⁵, Sungsu Kang^{2,3}, Tae Yoon Lee¹, Chihwan An¹, Seung Hyup Lee¹, Jong Seok Lee⁵, Jungwon Park^{2,3,6,7*}, Seung Chul Chae^{1,8*}

¹Department of Physics Education, Seoul National University, Seoul 08826, Republic of Korea

²School of Chemical and Biological Engineering, Institute of Chemical Process, Seoul National University, Seoul 08826, Republic of Korea

³Center for Nanoparticle Research, Institute for Basic Science (IBS), Seoul 08826, Republic of Korea

⁴Samsung Advanced Institute of Technology, Samsung Electronics, Suwon-si, Gyeonggi-do 16678, Republic of Korea

⁵Department of Physics and Photon Science, Gwangju Institute of Science and Technology (GIST), Gwangju, 61005, Republic of Korea

⁶Institute of Engineering Research, College of Engineering, Seoul National University, Seoul 08826, Republic of Korea

⁷Advanced Institute of Convergence Technology, Seoul National University, Suwon-si, Gyeonggi-do 16229, Republic of Korea

⁸Center for Education Research, Seoul National University, Seoul 08826, Republic of Korea

The stabilization of intermediate polar phases in fluorite-structured oxides is critical for advancing ferroelectric and antiferroelectric applications. Here, we report the stabilization of epitaxial polar tetragonal (T) ZrO₂. Epitaxial Hf_xZr_{1-x}O₂ thin films (x = 1, 0.75, 0.5, 0.25, and 0) are synthesized on (001) yttria-stabilized zirconia (YSZ) substrates via atomic layer deposition. The emergence of the unprecedented polar T-ZrO₂ phase deviates from the expected phase transition from non-polar HfO₂ through ferroelectric Hf_{0.5}Zr_{0.5}O₂ to anti-ferroelectric Zr-rich Hf_xZr_{1-x}O₂. Second harmonic generation (SHG) measurements reveal unexpected inversion symmetry breaking in T-phase ZrO₂. High-resolution 4D-scanning transmission electron microscopy (4D-STEM) further confirms the presence of electric dipoles originating from off-centered oxygen displacements. These findings establish a pathway for the low-temperature epitaxial synthesis of HfO₂-ZrO₂ based materials and provide critical insight into the polar nature of T-phase ZrO₂.

Tunable ferroelectricity in epitaxial HfO₂/ZrO₂ multilayers via interface engineering

Hong Hyun Jeon¹, Sang Won An², Yu Bin Park², Ha-Seung Lee³, Wang Been Lee³, Taejoon Kim³, Jungmoon Lim¹, Yeongju Choi¹, Si Won Kim¹, Jong Il Jeong¹, SeungNam Cha¹, Young-Min Kim³, Sang Mo Yang², Woo Seok Choi¹

¹*Department of Physics, Sungkyunkwan University, Suwon 16419, Korea*

²*Department of Physics, Sogang University, Seoul 04107, Korea*

³*Department of Energy Science, Sungkyunkwan University, Suwon 16419, Korea*

HfO₂ and ZrO₂ have attracted significant attention due to their compatibility with silicon technologies, and their structural phases affecting the ferroelectric (FE) and antiferroelectric properties. Bulk HfO₂ and ZrO₂ have the centrosymmetric monoclinic (M-phase) and thus are not FE. However, thin films of HfO₂ and ZrO₂ may exhibit FE behavior when stabilized in the orthorhombic (O-phase) through strain, doping, or oxygen vacancies [1]. Previous studies have mainly focused on Hf_{0.5}Zr_{0.5}O₂ (HZO) thin films because Zr incorporation easily stabilizes the FE O-phase of HfO₂. However, in conventional HZO solid-solution films, the FE response tends to degrade with increasing thickness [2]. To overcome the limitation, several studies have reported interface effects in pure HfO₂ and ZrO₂ multilayer structures, in which the O-phase is promoted by increased phase transition energy barriers and suppression of the M-phase by lattice constraints [3]. On the other hand, many studies have used atomic layer deposition, which is generally not suitable for fabricating multilayers with atomic-scale interface control because it typically produces polycrystalline films [4].

To understand the role of interface in the FE properties, we fabricated ultrathin epitaxial HfO₂-ZrO₂ multilayer on (110)-oriented La_{0.7}Sr_{0.3}MnO₃/SrTiO₃ substrates by pulsed laser epitaxy (PLE) [5]. Using such heterostructures with well-defined interfaces, we were able to investigate the control of ferroelectricity through interface engineering. Interestingly, all epitaxial multilayers exhibited robust O-phase confirmed by X-ray diffraction and scanning transmission electron microscopy (STEM) analyses. Also, atomically well-defined interfaces were confirmed from STEM. Accordingly, strong FE behavior was observed from $P(E)$ hysteresis loop measurements and piezoresponse force microscopy. More importantly, ferroelectricity showed a tendency to be enhanced as the number of interfaces between HfO₂ and ZrO₂ layers decreased. This enhancement may originate from differences in the distribution of oxygen vacancies at the interfaces. We anticipate that these results will provide a new approach for precisely controlling ferroelectricity in epitaxial HfO₂ and ZrO₂ multilayers.

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First-Principles EELS spectrum analysis in defective HfO₂

Minche Jo¹, Jaejin Hwang¹, Jaekwang Lee^{1,*}

¹ *Pusan National University, Republic of Korea*

HfO₂ has attracted significant attention as a promising material for next-generation semiconductor chip applications due to its high-k dielectric properties, scalable ferroelectricity, and compatibility with CMOS processing. Various defects that directly influence its electrical characteristics are difficult to identify at the atomic scale; in particular, the identification of point defects such as oxygen vacancies and complex defect configurations remains a challenging task. Therefore, establishing fingerprints corresponding to specific defect structures is of critical importance.

In this study, we employed density functional theory (DFT) calculations to investigate the electronic structure and electron energy-loss spectroscopy (EELS) spectra of selected defect structures in HfO₂, and derived defect-specific fingerprints.

In this presentation, we will discuss the fingerprints of specific defects, as well as spectral features that enable differentiation according to defect type and concentration.

Electro-Chemo-Mechanical Coupling in $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ Ferroionic Heterostructures

Achilles Bergne¹, Alikin Denis², Vasiljevic Milica³, Zamudio-García Javier³, Landberg Megan⁴, Marrero Lopez David⁵, Huang Sizhao¹, Valbjørn Christensen Dennis¹, Pryds Nini¹, Kholkin Andrei⁶, Esposito Vincenzo¹

¹ Department of Energy Storage and Conversion, Denmark Technical University - Lyngby (Denmark)

² CICECO Aveiro Institute of Materials, Physics Department, Campus de Santiago, University of Aveiro, 3810-193 - Aveiro (Portugal)

³ Department of Energy Storage and Conversion, Denmark Technical University - Lyngby (Denmark)

⁴ MAX IV Laboratory, Lund University, 22100 - Lund (Sweden)

⁵ Universidad de Málaga, Dpto. de Física Aplicada I, 29071 - Málaga (Spain)

⁶ Institute of Solid State Physics, University of Latvia, Kengaraga Str. 8, Riga, LV-1063 - Riga (Latvia)

Ferroelectricity in $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ (HZO) arises from a complex polymorphic energy landscape in which a metastable orthorhombic phase competes with monoclinic and tetragonal phases, rendering its functional properties extremely sensitive to structural instabilities [1]. Recent advances have leveraged ionic vacancy-mediated mechanisms—either via redox interactions with the surrounding environment or through the design of ferroionic heterostructures—to improve ferroelectric performance [2]. In this work, we adopt the ferroionic heterostructure paradigm and demonstrate that dynamic oxygen-vacancy exchange at epitaxial interfaces enables an active coupling between ferroelectric and ionic layers [2]. Epitaxial heterostructures incorporating $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_{3-\delta}$ (LSMO), yttria-stabilized ZrO_2 (YSZ), and Gd-doped CeO_2 (CGO) exhibit strongly coupled electro-chemo-mechanical responses, manifested by ferroelectric diode behavior and subtle interfacial lattice distortions. In particular, epitaxial fluorite-fluorite interfaces function as effective vacancy-exchange gates that bias phase stability, enhance polarization, amplify the piezoelectric response, and suppress leakage currents, in stark contrast to electronically dominated perovskite-fluorite junctions [3]. These results reveal that ferroionic heterostructures support reciprocal, vacancy-driven dynamics, establishing them as a versatile platform for defect-engineered ferroelectricity and tunable functionalities in hafnia-based oxide systems [4].

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Correlating Surface Topography with Ferroelectric and Leakage Phenomena in Hafnia Thin Films

Anudeep Tullibilli¹, Kartick Biswas¹, Shubham Kumar Parate¹, Pavan Nukala¹

¹*Centre for Nano Science and Engineering (CeNSE), Indian Institute of Science, Bengaluru 560012, India*

Abstract

Unconventional ferroelectricity in doped hafnia systems is often attributed to the migration of oxygen vacancies [1]. The wake-up effect is also understood to result from phase transitions mediated by the migration and redistribution of oxygen vacancies. These phase transitions induce morphological changes owing to volume differences of different phases [2]. Understanding the morphological changes driven by oxygen vacancies, redistribution offers valuable insight into why some ferroelectric devices exhibit increased leakage currents. In this study, we investigated the influence of oxygen vacancies distribution on the ferroelectric properties of Ytria doped epitaxial thin films, and its correlation with surface topography and the wake-up effect using Scanning Probe Microscopy techniques. Ferroelectric samples after wakeup exhibit a uniform height contrast between the probed and pristine region owing to reversible motion of a 2D oxygen vacancy front with field. Leaky sample upon cycling show drastic changes in roughness due to dendritic formation of oxygen vacancies. The observed morphological differences upon biasing indicate different modes of oxygen vacancies migration and redistribution in a leaky and ferroelectric sample as shown in Figure 1.

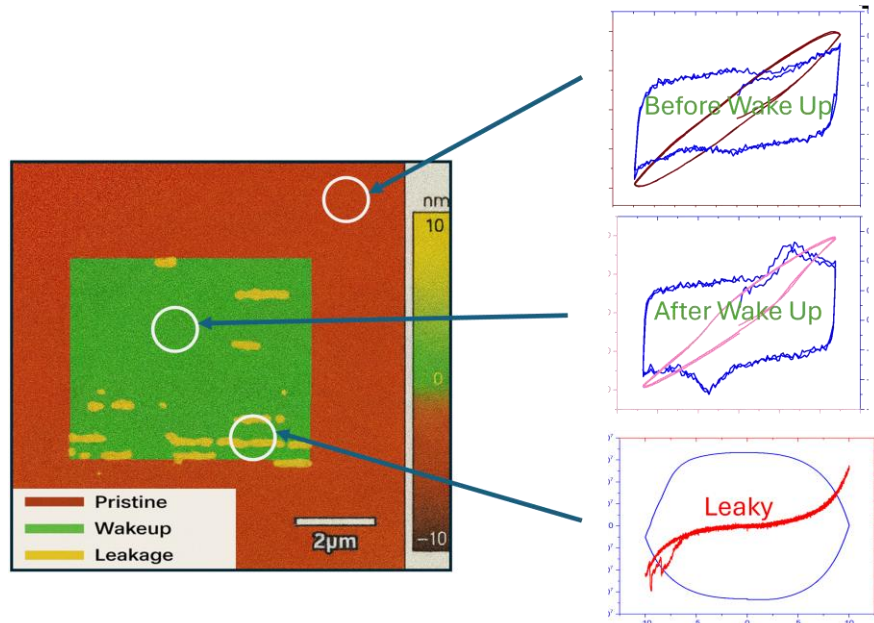


Figure 1: AFM topography image showing spatially distinct pristine, wake-up, and leakage regions identified after local electrical stressing. Image derived from experimental AFM data; colors and annotations added for visualization.

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Phase Transition Temperature Evolution in Ferroelectric Nanotubes with Different Wall Thicknesses via Electrical and Structural Characterization

Jin Kyu Han, Sunu Park, Ji Yong Hyun, and Sang Don Bu

Department of Physics, Jeonbuk National University, Jeonju 54896, Korea

Ferroelectric nanotubes exhibit unique physical properties arising from their reduced dimensionality, curved geometry, and enhanced surface-related effects, distinguishing them from conventional ferroelectric thin films and bulk ceramics. In ferroelectric systems, physical properties are strongly influenced by boundary conditions, including thickness, surface and interface energies, crystallographic symmetry, and geometrical confinement. These factors play important roles in determining key properties, including spontaneous polarization, dielectric response, piezoelectric behavior, and phase transition temperature. Owing to their hollow cylindrical structure, ferroelectric nanotubes are subjected to curvature-induced stress and surface tension, which have been theoretically predicted to enhance the phase transition temperature relative to two-dimensional thin films and three-dimensional bulk materials. Despite these theoretical predictions, experimental studies of the wall-thickness-dependent phase transition behavior of ferroelectric nanotubes remain limited, particularly with respect to combined electrical and structural characterization.

In this study, the phase transition temperature of PbTiO_3 (PTO) nanotubes with wall thicknesses ranging from approximately 7 to 70 nm is systematically investigated using temperature-dependent impedance measurements and high-temperature X-ray diffraction (HT-XRD) analysis. The nanotubes are fabricated using a template-assisted sol-gel process combined with spin-coating, enabling precise control of the nanotube wall thickness by adjusting solution concentration and coating cycles. The phase transition temperature is primarily determined from anomalies observed in the temperature-dependent impedance and dielectric responses, while HT-XRD measurements are performed to verify the corresponding structural transition. The results reveal that PTO nanotubes with a wall thickness of approximately 10 nm exhibit a significantly enhanced phase transition temperature of about 545 °C, which is notably higher than that of conventional PTO thin films. These findings provide experimental evidence that reduced wall thickness and curvature-related effects can stabilize the ferroelectric phase at elevated temperatures and demonstrate that impedance analysis is an effective approach for probing phase transitions in ferroelectric nanotube systems.

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Substrate-Controlled Ferroelectric Domain Structure in Epitaxial BaTiO₃: A Real-Space View of the Oxygen Octahedral Network

Roham Talei^{1,†}, Yupeng Chen^{2,†}, Bruce A. Davidson², Ke Zou², Nicolas Bonmassar¹

¹ *University of Stuttgart, Institute of Materials Science, Department for Materials Physics, Heisenbergstr. 3, 70569, Stuttgart, Germany*

² *Quantum Matter Institute, Department of Physics and Astronomy, University of British Columbia, 2355 East Mall, Vancouver, BC V6T 1Z4, Canada*

[†] *These authors contributed equally to this work.*

BaTiO₃ (BTO) has one of the largest known Pockels coefficients the refractive-index change under an electric field and grows epitaxially on silicon via a SrTiO₃ (STO) buffer, making it promising for electro-optic modulators and switches in silicon photonics [1]. Its usefulness is governed by the ferroelectric domain structure: oppositely oriented domains contribute with opposite sign, so an uncontrolled multi-domain film exhibits a vanishing effective Pockels coefficient [2], and the optimal orientation depends on device geometry [3]. Controlling the domain configuration is therefore the decisive challenge, and the substrate is the most accessible lever.

We compare ~100 nm BTO films grown by molecular beam epitaxy (MBE) on STO ($a = 3.905$ Å, ≈ -2.2 % misfit) and LSAT [(LaAlO₃)_{0.3}(Sr₂TaAlO₆)_{0.7}, $a = 3.868$ Å, ≈ -3.1 % misfit]. Both impose compressive in-plane strain that nominally favours c-domains, but at this thickness the films relax beyond the critical thickness for single-domain growth. X-ray diffraction shows STO-grown films forming a mixed a/c domain state through ferroelastic relaxation, whereas LSAT-grown films stay mainly c-domain oriented with a markedly narrower rocking curve, despite the larger misfit. That the higher-misfit substrate yields higher domain purity and better crystallinity contradicts the expectation that larger misfit degrades the film. Since the polarization originates in Ti off-centering within its oxygen octahedron, we hypothesize that the relaxation pathway is governed by the local TiO₆ tilt field set by interfacial bonding and thermal-expansion mismatch on cooldown and that broader tilt heterogeneity nucleates the a-domains on STO. Conventional diffraction averages over the lattice and is blind to this nanoscale tilt distribution.

Resolving it requires a real-space probe sensitive to both the Ti positions and the oxygen sublattice at atomic resolution. We therefore image both films by aberration-corrected scanning transmission electron microscopy (STEM), combining high-angle annular dark-field (HAADF) imaging of the heavy Ba and Ti columns with integrated differential phase contrast (iDPC) imaging of the light oxygen columns, giving direct access to the octahedral tilt geometry. From these images we build unit-cell-resolved Ti-displacement maps, converting atomic positions into local polarization vectors within domains and at domain walls, revealing features beyond the reach of diffraction or surface characterization [4]. By correlating the tilt and defect distributions with the domain ordering on STO and LSAT, we aim to establish the microscopic link between defects, octahedral tilts, and a/c domain formation, identifying TiO₆ tilt as a design knob for predictable domain structures and hence a reproducible electro-optic response in BTO on silicon.

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Unconventional magnon spin polarization enabled by ferroelectric domain engineering in multiferroic oxide

Yuhan Liang^{1,2}, Daniel C. Ralph², Tianxiang Nan¹

¹ *School of Integrated Circuits and Beijing National Research Center for Information Science and Technology (BNRist), Tsinghua University, Beijing 100084, China*

² *Department of Physics, Cornell University, Ithaca, NY 14853, USA*

Spin current provides an energy-efficient approach for manipulating magnetization, when its spin polarization aligns with the magnetization direction^[1]. However, conventional spin-source materials possess high crystalline symmetry, restricting spin polarization to be orthogonal to both spin and charge current directions^[2]. Here, we overcome this limitation by utilizing the concept of magnon-mediated spin-orbit torque through integration of the insulating multiferroic bismuth ferrite with a conventional spin-source material^[3]. We observe that spin polarization generated by conventional spin-source material can excite unconventional magnon polarization due to the interplay between cycloidal antiferromagnetic order and the ferroelectric domain structure in bismuth ferrite. This produces an unconventional magnon torque that allows deterministic, field-free switching of in-plane magnetization collinear with the current direction, unattainable with conventional spin-source materials. Our results establish multiferroic-based heterostructure as a symmetry-engineered magnon spin source, paving the way for low-power spintronic devices.

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Effect of PbTiO₃ Nanotube Wall Thickness on Piezoelectric Nanogenerator Performance

Sunu Park¹, Jin Kyu Han¹, and Sang Don Bu^{1,*}

¹Department of Physics, Jeonbuk National University, Jeonju 54896, Korea

With the rapid growth of IoT and wearable electronics, energy harvesting technologies have attracted considerable attention, among which piezoelectric nanogenerators (PENGs) are promising self-powered energy sources that convert mechanical energy into electrical energy.

To improve the power generation efficiency, materials such as nanowires, nanoparticles, and nanotubes, as well as structural modifications of the generator, have been widely employed.

Among them, nanotubes, which have a unique hollow cylindrical structure, exhibit characteristics different from those of two-dimensional films or three-dimensional bulk structures.

According to the Ginzburg–Landau–Devonshire theory, surface tension and size effects in ferroelectric nanotubes can enhance their phase transition temperature, polarization, and piezoelectric properties as the wall thickness decreases. In particular, the piezoelectricity of nanotubes with a specific wall thickness is known to be maximized.

In this study, we fabricated piezoelectric nanogenerators based on ferroelectric nanotubes and investigated their performance as a function of nanotube wall thickness.

PbTiO₃ (PTO) nanotubes (PTO-NTs) with a length of 1 μm and a diameter of 400 nm were fabricated by spin-coating a sol–gel-derived PTO solution onto anodic aluminum oxide (AAO) membranes. The wall thickness of the PTO-NTs was controlled from 10 to 70 nm by varying the number of spin-coating cycles. The fabricated PTO-NTs were confirmed to possess a tetragonal perovskite phase by X-ray diffraction (XRD) and Raman spectroscopy. Subsequently, PENG devices were fabricated using the PTO-NTs, and their electrical output performance was evaluated under applied pressure.

This research was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIT) (No. RS-2025-02217073) and the financial support of the Global-Learning & Academic research institution for Master's·PhD students, and Postdocs (LAMP) Program of the National Research Foundation of Korea (NRF) grant funded by the Ministry of Education (Grant No. RS-2024-00443714).

Corresponding author : sbu@jbnu.ac.kr

Hydrothermal Synthesis of Tetragonal BaTiO₃ Nanotubes for Efficient Piezoelectric Nanogenerators

Jiyong Hyun¹, Sunu Park¹, Jin Kyu Han¹, Sang Don Bu^{1*}

¹ *Department of Physics, Jeonbuk National University, Republic of Korea*

The rapid advancement of wearable electronics and the internet of Things (IoT) has triggered a critical demand for independent, self-powered energy sources. Consequently, piezoelectric nanogenerators (PENGs) have emerged as highly promising next generation energy harvesting technologies, capable of directly converting ambient mechanical stimuli into electrical power. While conventional PENGs predominantly utilize materials such as ZnO, PZT, and PTO, their practical implementation in wearable devices is severely hindered by limited piezoelectric performance or the environmental and health hazards associated with toxic lead. To address these critical bottlenecks, this study introduces a high-performance, eco-friendly PENG employing barium titanate (BaTiO₃) a quintessential lead-free ferroelectric material. Specially, to maximize both energy conversion efficiency and mechanical flexibility, one BaTiO₃ nanotubes (BTO-NTs) were directly synthesized on anodized Ti wire.

In this work, the Ti wire was initially anodized and subsequently subjected to hydrothermal synthesis at 200°C for varying reaction times (6 to 48 hours) to systematically investigate the time-dependent morphological evolution and the corresponding piezoelectric performance. Scanning electron microscopy (SEM) confirmed the successful formation of TiO₂, BTO nanotubes, with both the outer diameter and wall thickness increasing proportionally with the hydrothermal reaction time. Furthermore, X-ray diffraction (XRD) analysis revealed that prolonged synthesis induced a distinct splitting of the (200) and (002) peaks, indicating significantly enhanced tetragonality of the BTO phase. X-ray photoelectron spectroscopy (XPS) further verified the chemical composition and bonding states of the synthesized BTO-NTs.

For device integration, indium tin oxide (ITO) and the Ti wire core served as the bottom and top electrodes, respectively. While the active BTO-NT layer was encapsulated in polydimethylsiloxane (PDMS) to ensure electrical insulation and robust mechanical durability. To maximize the piezoelectric output, the device was poled under DC bias of 1000 V for 6 hours. Subsequent electrical measurements using an oscilloscope demonstrated clear polarity switching, confirming successful polarization and confirming that the generated output originates strictly from the intrinsic piezoelectric effect. By systematically measuring the output voltage and current across varying external load-resistances, the device's maximum power density was effectively extracted. Ultimately, the BTO-NT based flexible PENG developed in this study simultaneously achieves superior energy generation and eco-friendliness, highlighting its tremendous potential as a sustainable, independent power source for next-generation wearable electronics and self-powered IoT platforms.

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Atomic-scale characterisation of multiferroic oxide Aurivillius thin films using Transmission Electron Microscopy and Atom Probe Tomography

Geri Topore¹, Lynette Keeney², Quentin Ramasse^{3,4}, Baptiste Gault⁵, Shelly Conroy¹

¹ *Department of Materials, Imperial College London, UK*

² *Tyndall National Institute, University College Cork, Cork, Ireland*

³ *SuperSTEM Laboratory, SciTech Daresbury Campus, Daresbury, UK*

⁴ *School of Chemical Process Engineering, University of Leeds, Leeds, UK*

⁵ *Univ Rouen Normandie, CNRS, INSA Rouen Normandie, Groupe de Physique des Matériaux*

Magnetoelectric multiferroic materials offer significant potential for low-power computer memory and storage devices due to their coupled ferromagnetism and ferroelectricity [1]. Naturally layered Aurivillius-phase $\text{Bi}_6\text{Ti}_x\text{Fe}_y\text{Mn}_z\text{O}_{18}$ (B6TFMO) thin films have shown to exhibit such behaviour at room temperature as a single-phase material [2], making it a promising system to study the fundamental physics surrounding the mechanisms of multiferroicity and the manipulation of coupled domains, as well as higher order topologies including polar vortices that have been shown to emerge around defects in this material. In this work, we utilise scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) to show not only the preferential partitioning of B-site cations (Ti, Mn, Fe) in the perovskite superlattice, but how the crystal field splitting and valence of these cations changes within perovskite layers at atomic resolution by analysing the fine structure in EEL spectrum images. We relate such changes to strain and the presence of octahedral tilting, measured directly from atomic-resolution integrated differential phase contrast (iDPC) imaging. While techniques like STEM and EELS provide critical insights, they are fundamentally limited to a 2D-projected view of inherently 3D features. To overcome this limitation for compositional analysis, we employ Atom Probe Tomography (APT) as a complementary technique to analyse the 3D chemistry changes in small nanometre-sized volumes, providing insight into the segregation of elements of interest around defects like out-of-phase boundaries (OPBs) where we have observed polar vortices emerge.

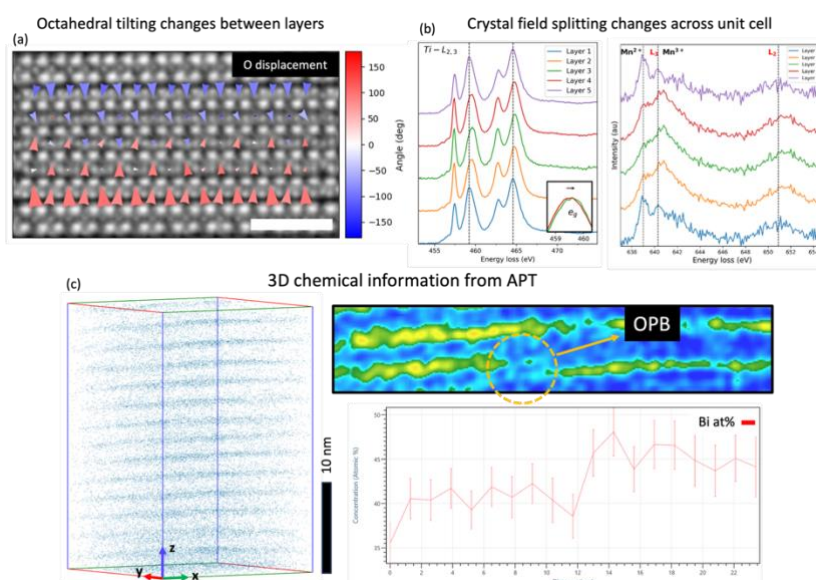


Figure 1: (a) iDPC image showing octahedral tilting across a unit cell. Scalebar 1 nm. (b) EEL spectra showing how the fine structure changes across the unit cell for Ti and Mn. (c) APT analysis showing a 3D reconstructed volume across the film thickness and Bi concentration around an OPB.

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Electric-Field-induced Weakly Ferromagnetic Domain Change by Ferroelectric Topological Domain Switching in Co-Substituted BiFeO₃ Nanodots

Koomok Lee^{1*}, Peter Meisenheimer², Paul Stevenson³, Yasuhito Nagase¹,
Kei Shigematsu^{1,4*}, Ramamoorthy Ramesh^{2,5}, Masaki Azuma^{1,4}

1. Institute of Integrated Research, Institute of Science Tokyo, Yokohama, Japan
2. Department of Materials Science and Engineering, University of California, Berkeley, USA
3. Department of Physics, Northeastern University, Boston, USA.
4. Kanagawa Institute of Industrial Science and Technology, Ebina, Japan
5. Department of Materials Science and Nanoengineering, Department of Physics and Astronomy, Rice University, Houston, USA

Perovskite BiFe_{0.9}Co_{0.1}O₃ (BFCO) is a room temperature multiferroic material where ferroelectric polarization is coupled to spontaneous magnetization[1]. Through electric field control of magnetization reversal of BFCO, it is highly promising for low-power-consumption, non-volatile, magnetic memory application.

In this study, we investigate the electric field manipulation of both ferroelectric and ferromagnetic domain changes in BFCO nanodots fabricated by pulsed laser deposition[2]. Figure 1a shows the topography of BFCO nanodots with diameter of 190 nm. Ferroelectric topological center-quadrant domain was observed by piezoresponse force microscopy where downward polarization pointing inward in as-grown state (Fig. 1b). After electric poling, polarization switched to upward to outward direction. Magnetic domain in the same dot was mapped using nitrogen-vacancy center magnetometry (Fig. 1c). The reversal of magnetization by electric poling was confirmed and its magnetization directions are revealed in M_{dot} .

This study demonstrates the electric field control of magnetization in multiferroic domains paving the way for the design of future voltage-write magnetic memory[3].

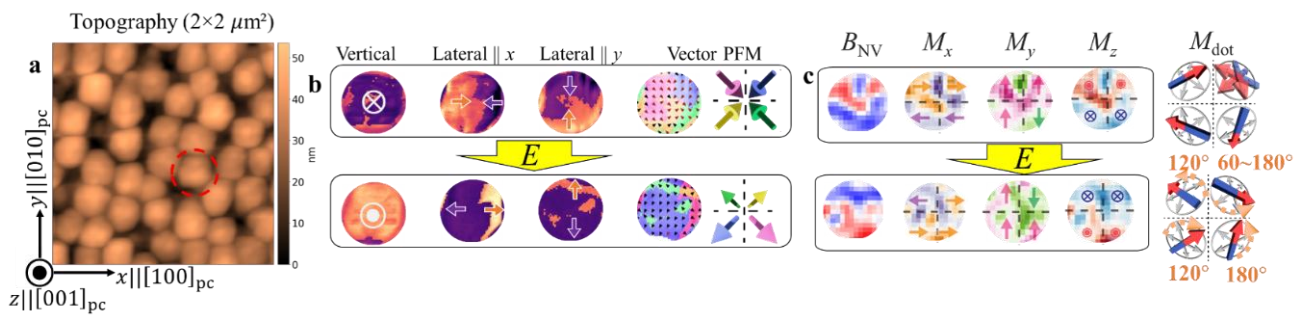


Figure 1 **a**, Topographic image of 190 nm BFCO nanodots in 2×2 μm² region. **b**, Ferroelectric topological domain change from downward / center-convergent to upward / center-divergent state by electric poling. **c**, Magnetic stray field (B_{NV}) and magnetization (M) along x , y , and z directions in ferromagnetic domain before and after electric poling. M_{dot} illustrates the possible M directions coupled to polarization in each quadrant domain.

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RF-magnetron-Sputtered Sc:TiO₂ Thin Films Exhibiting High-ON/OFF ratio Bipolar Resistive Switching Properties

T. Yabuki¹, A. Maekawa¹, S Daisuke², H Kumigashira², and Tohru Higuchi¹

¹ Department of Applied Physics, Tokyo University of Science, Tokyo 125-8585, Japan

² Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan

Resistive random access memory (ReRAM), a promising next-generation nonvolatile memory, features a simple structure and fast switching capabilities, however, reducing power consumption associated with excessive current flow in the low-resistance state remains a critical challenge for practical application. To address this issue, the utilization of wide-bandgap semiconductors, which inherently have low carrier concentrations and can suppress operating current, is considered effective.

In this study, to develop ReRAM devices with low power consumption and a high resistance switching ratio, anatase TiO₂ thin films doped with 1% Sc (Sc:TiO₂) were fabricated, and their structural and resistive switching properties were evaluated. By substituting Sc³⁺ at the Ti⁴⁺ sites, oxygen vacancies were intentionally generated. A device structure (Pt/Sc:TiO₂/Nb:TiO₂/Pt) incorporating a highly conductive Nb-doped TiO₂ layer as an oxygen reservoir was constructed using RF magnetron sputtering.

Evaluations using X-ray absorption spectroscopy (XAS) and X-ray photoemission spectroscopy (XPS) confirmed the successful incorporation of Sc and a mixed valence state of Ti (Ti³⁺/Ti⁴⁺), revealing that Sc doping generated a large amount of oxygen vacancies (1.32%). Furthermore, the optical bandgap was estimated to be 3.78 eV using a Tauc plot derived from optical transmittance measurements. Current-voltage (I-V) measurements of the device exhibited a stable, continuous bipolar switching behavior with a set voltage of around 2.7 V, which is attributed to the modulation of the Schottky barrier. The device recorded an excellent ON/OFF ratio of 124.76 at a read voltage of 0.5 V and a high-speed response on the nanosecond order. This study demonstrates that the promotion of oxygen vacancy formation and the partial reduction of Ti induced by Sc doping lead to superior resistive switching properties, providing fundamental insights toward the development of low-power TiO₂-based ReRAM devices.

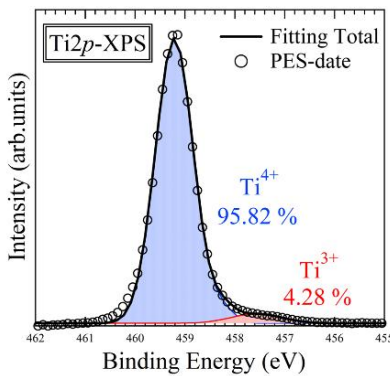


Figure 1

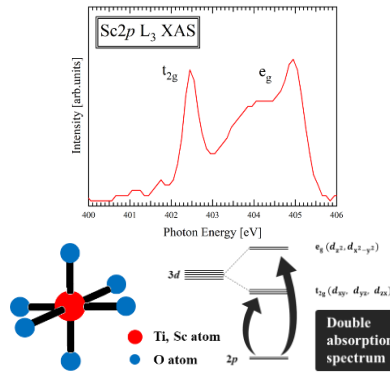


Figure 2

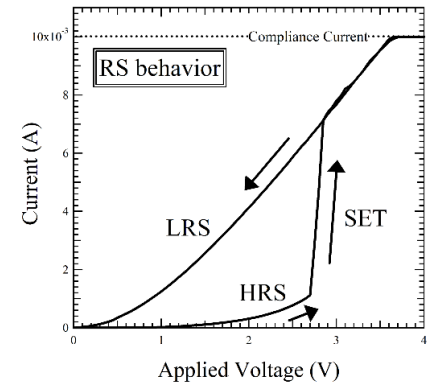


Figure 3

Figure 1: Ti 2p XPS Spectrum and Quantification of Oxygen Vacancies, Figure 2: Sc 2p XAS Spectrum and Schematic diagram of crystal-field-induced spectral splitting, Figure 3: Resistive switching behavior of M-I-M capacitor type device

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Antiferroelectric-like polarization reversal induced by ordered polar vortices

Tom Zander¹, Tobias Truschies¹, Lei Jin², Abhinav Raina³, Lide Yao⁴, Oana-Andreea Condurache⁵, Le Van Lich⁶, Tien-Nhan Nguyen⁷, Sebastiaan van Dijken⁸, Paul H. M. van Loosdrecht¹ and Ionela Lindfors-Vrejoiu¹

¹*II. Physikalisches Institut, Universität zu Köln, 50937 Köln, Germany*

²*Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany*

³*Institute for Light and Matter, Universität zu Köln, 50939 Köln, Germany*

⁴*OtaNano-Nanomicroscopy Center, Aalto University, P.O. Box 15100, FI-00076 Aalto, Finland*

⁵*Laboratoire Structures, Propriétés et Modélisation des Solides (SPMS), Université Paris-Saclay, CentraleSupélec, Gif-sur-Yvette, France*

⁶*Center for Materials Innovation and Technology, Vin University, Hanoi, Vietnam*

⁷*School of Materials Science and Engineering, Hanoi University of Science and Technology, No.1 Dai Co Viet Street, Hanoi, Vietnam*

⁸*Department of Applied Physics, Aalto University School of Science, P.O. Box 15100, FI-00076 Aalto*

In heterostructures of ferroelectric and paraelectric perovskite oxides polar phases such as vortices and skyrmions of few nanometer size were observed and are stable at room temperature [1]. Antiferroelectrics, which have ordered antiparallel dipoles, can transition to a polar (parallel-dipole) state under applied field. Due to their characteristic double-loop in the polarization-electric field curve, AFE capacitors exhibit high energy storage densities and efficiency [2]. Alternatives to bulk antiferroelectrics, like PbZrO_3 , are worth searching for, especially in thin film configurations [3]. Similar polarization reversal response as to antiferroelectric films, resulting in polarization-electric field double-loops, can be obtained from heterostructures in which the vortex state dominates. These vortex domains can be manipulated by an electric field and the polarization-field hysteresis loops resemble antiferroelectric behavior [3]. Here, the polarization reversal in epitaxial heterostructures of trilayer $\text{SrTiO}_3/\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3/\text{SrTiO}_3$ is investigated. The heterostructures are grown under biaxial in-plane tensile strain on $\text{GdScO}_3(110)$ substrates. In such heterostructures a vortex lattice of ordered pairs of opposite winding and about 9 nm periodicity forms as observed by transmission electron microscopy (Fig. c)) and X-ray diffraction (Fig. d)). The asymmetric biaxial tensile strain strongly affects the ferroelectric properties. The in-plane polarization hysteresis loops of STO/PZT/STO trilayers measured at room temperature show double-loops (Fig. b)), resembling the polarization hysteresis loops of antiferroelectric materials. Their origin is the response of the ordered antipolar nanodomains of the trilayers, which can be switched to a dipolar wave-like polar state under an electric field [4].

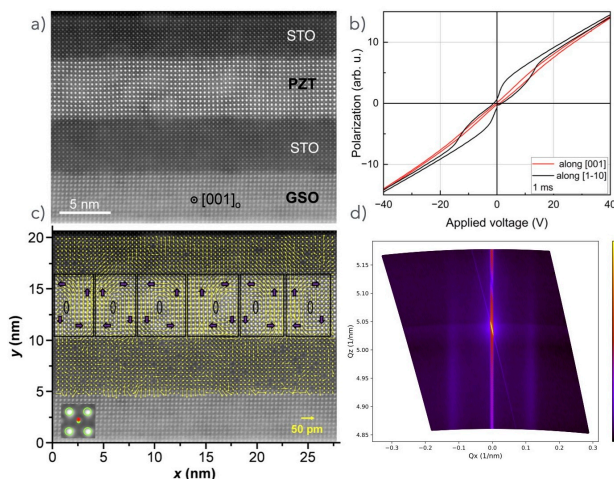


Figure 1: a) Cross-sectional transmission electron microscope picture. b) Polarization hysteresis loops along two crystallographic in-plane directions. c) Atomic displacement map and d) Reciprocal space map around the (002)_{pc} substrate Bragg peak.

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Giant shift current in ferroelectric halide perovskite CsGeI₃ epitaxial thin films

Masao Nakamura^{1,2,3}, Koma Miki^{4,5}, Asahi Yamada¹, Gurvan Bosser², Kiyohiro Adachi², Daisuke Hashizume², Naoki Ogawa^{2,3}, Satoshi Okamoto^{3,6}, Yoshinori Tokura^{2,3,7}, and Masashi Kawasaki^{2,3}

¹*Department of Physics, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan*

²*RIKEN Center for Emergent Matter Science (CEMS), Wako 351-0198, Japan*

³*RIKEN Baton Zone Program (BZP), Wako 351-0198, Japan*

⁴*Department of Applied Physics and Quantum-Phase Electronics Center (QPEC), The University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan*

⁵*Institute of Industrial Science, The University of Tokyo, Meguro-ku, Tokyo 153-8505, Japan*

⁶*Sumitomo Chemical Co., Ltd, Japan, Chuo-ku, Tokyo, 103-6020, Japan*

⁷*Tokyo College, The University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan*

Email: masao.nakamura.b8@tohoku.ac.jp

Halide perovskites have recently attracted considerable attention as promising materials for solar cells. Among them, Ge-based halide perovskites such as CsGeI₃ exhibit robust ferroelectricity with a large spontaneous polarization exceeding 20 $\mu\text{C}/\text{cm}^2$ at room temperature [1], together with strong visible-light absorption due to their direct bandgap of 1.6 eV (Fig. 1(a)). Furthermore, CsGeI₃ is expected to exhibit strong nonlinear optical responses owing to its large polarization. In particular, a significant enhancement of the shift current, a nonlinear photocurrent governed by the quantum geometric properties of Bloch wavefunctions, has been theoretically predicted in this compound [2]. However, its photovoltaic and nonlinear properties have remained largely unexplored because of the difficulty in fabricating high-quality thin films.

Recently, we succeeded in growing high-quality CsGeI₃ thin films on BaF₂(001) substrates by employing molecular beam epitaxy [3]. The films exhibit a clear zero-bias photocurrent above the bandgap with characteristic signatures of shift current, including spectral sign reversals, light-polarization dependence, and reversible electric-field modulation associated with switchable ferroelectric polarization (Fig. 1(b)). Remarkably, the magnitude of the shift current, evaluated using the Glass coefficient, is the highest reported among known compounds (Fig. 1(c)). The results demonstrate the potential of ferroelectric halide perovskites as a platform for high-performance photovoltaic and nonlinear optoelectronic applications.

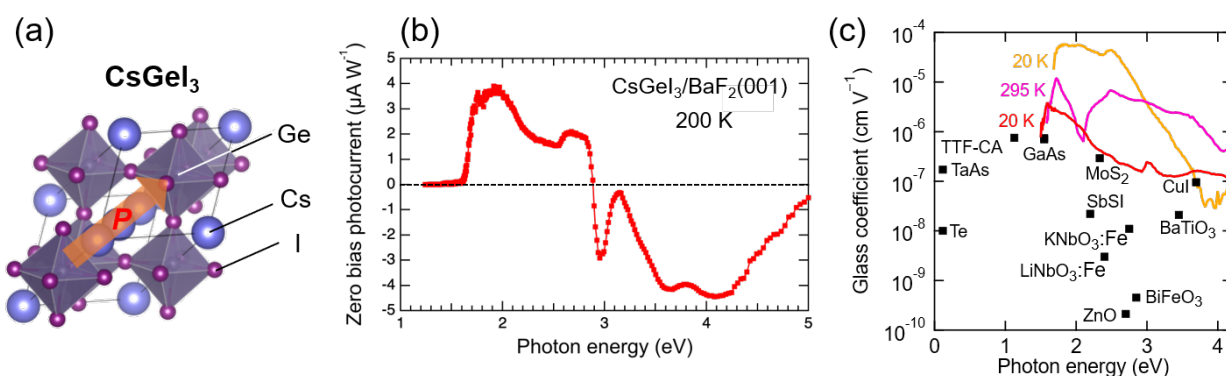


Figure 1: (a) Schematic crystal structure of CsGeI₃. (b) Excitation spectrum of zero-bias photocurrent in a CsGeI₃/BaF₂(001) thin film. (c) Comparison of shift-current response between CsGeI₃ and other reported compounds.

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Expanding the Polarization Window of Epitaxial $\text{Hf}_x\text{Zr}_{1-x}\text{O}_2$ through an Unconventional Ferroelectric Switching Pathway

Chihwan An¹, Hamoon Fahrivandi², Dongmin Kim³, Baekjune Kang⁴, Pawan Kumar², Tae Yoon Lee¹, Jung Woo Cho¹, Seung Hyup Lee¹, Changhee Sohn⁴, Jungwon Park³, Jun Hee Lee² and Seung Chul Chae¹

¹ Department of Physics Education, Seoul National University, Seoul 08826, Republic of Korea

² Department of Energy Engineering, School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology, Ulsan 44919, Republic of Korea

³ Department of Chemical and Biological Engineering, Seoul National University, Seoul 08826, Republic of Korea

⁴ Department of Physics, Ulsan National Institute of Science and Technology, Ulsan 44919, Republic of Korea

Ferroelectric HfO_2 -based materials offer a promising candidate for integrating ferroelectric functionality into modern electronic devices owing to their compatibility with established CMOS technology. However, previous theoretical and experimental studies have suggested that the intrinsic remanent polarization (P_r) of HfO_2 -based ferroelectrics is limited to approximately $60 \mu\text{C}/\text{cm}^2$. This apparent ceiling restricts the accessible polarization window, requiring higher operating voltages to achieve reliable state separation under practical noise and variability conditions. Here, we report strongly enhanced polarization in epitaxial $\text{Hf}_x\text{Zr}_{1-x}\text{O}_2$ (HZO) thin films ($x = 0.50, 0.56, 0.60, 0.66$) grown by atomic layer deposition on (001)-oriented yttria-stabilized zirconia (YSZ) substrates. X-ray diffraction analyses confirm that the HZO films are epitaxially aligned with the underlying YSZ substrate. Precise electrical measurements reveal an out-of-plane P_r as high as $120 \mu\text{C}/\text{cm}^2$ at room temperature, exceeding the previously reported polarization limit of HfO_2 -based ferroelectrics. This unprecedented polarization suggests the activation of extended oxygen-ion displacements associated with an unconventional switching pathway distinct from the widely accepted polarization-switching mechanism. These findings expand the accessible ferroelectric response of HfO_2 -based materials and provide new opportunities for enhancing the performance of ferroelectric electronic devices.

Enhanced Rashba Spin-Orbit Interactions in polar oxide 2DEGs

Janine Gückelhorn¹, Sergi Plana-Ruiz^{2,3}, Gyanendra Singh¹, Saul Estandia

Rodriguez¹, Roger Guzman¹, Kapil Gupta³, Fernando Gallego⁴, Luis M. Vicente-Arche⁴, Joaquim

Portillo⁵, Thanos Galanis⁵, Manuel Bibes⁴, Jaume Gázquez¹, and Gervasi Herranz¹

¹ *Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus de la UAB, Bellaterra, Catalonia, Spain.*

² *Unitat de Cristal·lografia i Mineralogia, Departament de Geologia, Universitat Autònoma de Barcelona, Cerdanyola del Vallès, Catalonia, Spain.*

³ *Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus de la UAB, Bellaterra, Catalonia, Spain.*

⁴ *Laboratoire Albert Fert, CNRS, Thales, Université Paris Saclay, Palaiseau, France.*

⁵ *NanoMEGAS SPRL, Rue Èmile Claus 49 bte 9, Brussels, Belgium.*

Two-dimensional electron gases (2DEGs) at oxide interfaces exhibit strong Rashba spin-orbit coupling (SOC), originating from broken inversion symmetry and the associated built-in electric field. Density functional theory predicts that Rashba interactions are enhanced in materials with high dielectric polarizability, pointing to two possible microscopic mechanisms underlying Rashba SOC: polar lattice displacements and orbital polarization induced by the interfacial electric field [1]. In general, the Rashba SOC strength can be enhanced by applying external electric fields. The critical role of lattice distortions was first demonstrated experimentally by modulating Ti–O lattice polarization via a thin LaFeO₃ spacer between SrTiO₃ and LaAlO₃, which enhances the interfacial Rashba SOC [2]. However, to date, the specific role of the electric field has remained unexplored.

Here, we use a fundamentally different approach by forming the 2DEGs at AlO_x/Sr_{1-x}Ca_xTiO₃ interfaces, demonstrating a substantial enhancement of the Rashba coefficient driven by the increased dielectric polarizability of the substrate [3]. Substituting a small fraction of Sr with Ca in SrTiO₃ ($x = 0.01, 0.02, 0.03$) promotes ferroelectricity and increases the dielectric constant, reflecting its large polarizability at low temperatures. Increasing the Ca content, we observe a corresponding increase in Rashba spin-orbit coupling. Interestingly, for higher Ca content, the Rashba coefficient α_R becomes nearly independent of the carrier density or applied electric fields, respectively. Room-temperature atomic-scale structural and electrostatic analysis shows the presence of both lattice distortions and built-in electric fields that vary only weakly with Ca content. Though these observations are well above the ferroelectric transition, leaving the mechanism prevailing at low temperatures open, they nevertheless indicate that dielectric polarizability is a crucial parameter governing Rashba spin-orbit coupling at oxide interfaces and highlight dielectric engineering as a promising route for tuning spin-orbit interactions in oxide electronics.

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Toward Non-Volatile Ferroelectric Spin-Orbit Devices: A Room-Temperature 2DEG in STO/PZT Heterostructures

Thomas Buttiens^{1,2}, Anouk Goossens¹, Luis M. Vicente Arche¹, Stephane Chiroli¹, Marko Kuveždić¹, Davide Cammilleri², Maxime Vallet³, Brahim Dkhil³, Alexandre Gloter⁴, Thomas Maroutian², Manuel Bibes¹

¹Laboratoire Albert Fert, CNRS, Thales, Université Paris-Saclay, Palaiseau (France)

²Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Saclay, Palaiseau (France)

³Université Paris-Saclay, CentraleSupélec, CNRS, Laboratoire SPMS, 91190 Gif-sur-Yvette (France)

⁴Laboratoire de Physique des Solides, CNRS, Université Paris-Saclay, Orsay (France)

The ever-growing power consumption of today's electronics drives the need for emerging device concepts. Oxide-based materials are particularly promising building blocks, as they can host and combine a wide variety of functional properties (e.g., magnetism, ferroelectricity, spin-orbit coupling). Our approach targets logic-in-memory computing with non-volatile operation by combining ferroelectricity and spin-orbit coupling in ferroelectric spin-orbit (FESO) devices [1]. A key requirement for FESO devices is the ferroelectric control of efficient spin-charge conversion, which has so far been demonstrated only at cryogenic temperatures, in two-dimensional electron gases (2DEGs) formed at the surface of bulk SrTiO₃ (STO).

Here, we present an attempt to adapt a widely studied and industrially available ferroelectric material, Pb(Zr,Ti)O₃ (PZT), in the form of thin films for spin-orbitronics. Theoretical studies reveal a Rashba spin-orbit coupling character in the closely related compound PbTiO₃, potentially suitable for FESO devices [2]. We investigate the formation of a 2DEG at the PZT surface by depositing a reducing metal overlayer. The 2DEG formation is then studied by X-ray photoemission spectroscopy (XPS), and its magnetotransport properties are investigated. However, we encounter an inherent challenge at the PZT interface arising from lead interdiffusion: although the interface is conducting at room temperature, it exhibits low mobility and non-metallic behavior that renders it unsuitable for spin-charge conversion. We circumvent this issue by growing an additional STO ultrathin overlayer on top of the PZT, which stabilizes a 2DEG at the sample surface, combining spin-orbit coupling from the STO 2DEG and room-temperature ferroelectric control from the underlying PZT. Its properties are confirmed by XPS, magnetotransport measurements, P-E loop measurements, spin-pumping experiments, and scanning transmission electron microscopy (STEM). Finally, a patterning strategy to effectively study the ferroelectric tuning of transport and spin-orbit properties will also be presented. These results establish STO-capped PZT heterostructures as a viable room-temperature platform for FESO devices, opening a path toward ferroelectric control of spin-charge conversion and emerging non-volatile electronic devices.

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Interfacial polarity control of anomalous Hall effect in $\text{LaCoO}_3\text{-SrIrO}_3$ heterostructures

Yeonjae Lee¹, Chanyoung Shin², Youngdo Kim¹, Jinyoung Kim¹, Jaeung Lee¹, Miyoung Kim², Sangjae Lee³, Changyoung Kim¹

¹ Department of Physics and Astronomy, Seoul National University, Seoul, Korea

² Department of Materials Science and Engineering, Seoul National University, Seoul, Korea

³ The Research Institute of Basic Science, Seoul National University, Seoul, Korea

Interfacial polarity in oxide heterostructures can drive robust charge redistribution across the interface, yet its role in tailoring topological transport phenomena like the anomalous Hall effect (AHE) remains largely unexplored. Here, we control the sign of polarity-induced charge in $\text{SrIrO}_3\text{-LaCoO}_3$ interfaces by independently engineering the layer stacking sequence and the atomic termination. In-situ angle-resolved photoemission spectroscopy directly confirms the sign reversal of the induced charge in SrIrO_3 through the contrasting electronic band shifts between different interfacial configurations. Magneto-transport measurements across a systematic matrix of four distinct heterostructures reveal that the AHE can be cleanly toggled on and off: it emerges exclusively under electron-doped conditions, while remaining suppressed for hole-doped SrIrO_3 . Our results establish that this response originates from an intrinsic, Berry-curvature-driven mechanism governed by polarity-controlled band filling of SrIrO_3 . These findings shift interfacial polarity from a passive structural consequence to an active design variable for engineering topological quantum states in oxide heterostructures.

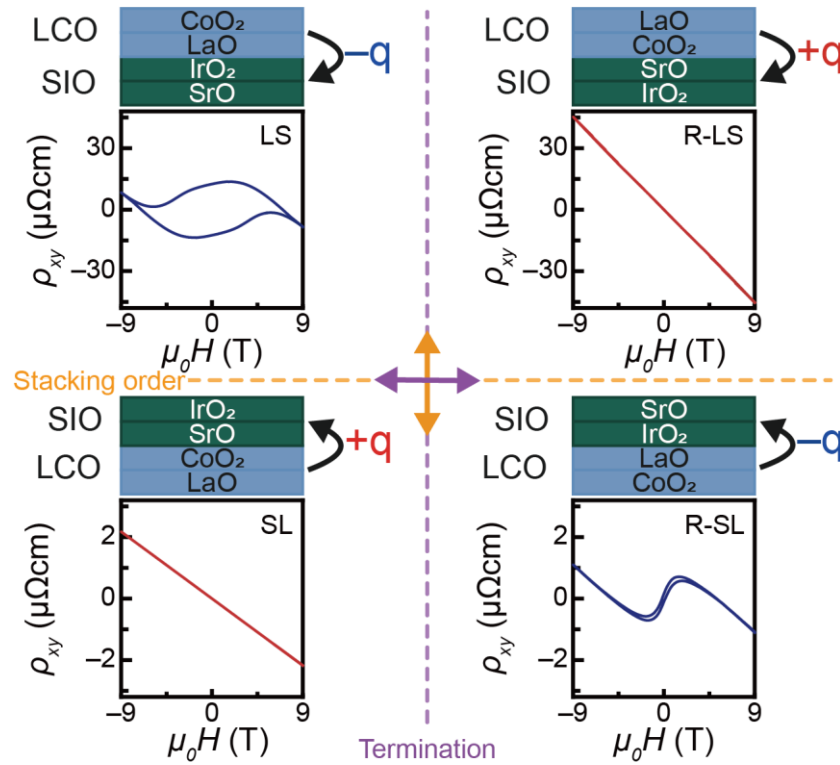


Figure 1: Schematic summary of the interfacial polarity engineering matrix and emergent topological transport

Room-temperature spin-polarized transport in SrTiO₃-based two-dimensional electron gas

Tatsuro Endo,¹ Keita Hidaka,¹ Minori Osaki,¹ Masaaki Tanaka^{1,2,3} and Shinobu Ohya^{1,2,3}

¹*Department of Electrical Engineering and Information Systems, The University of Tokyo*

²*Center for Spintronics Research Network, The University of Tokyo*

³*NanoQuine, The University of Tokyo*

The discovery of two-dimensional electron gas (2DEG) in the transition-metal perovskite oxide SrTiO₃ (STO) has stimulated intensive studies of this system. These studies have revealed various intriguing properties, including functionalities relevant to spintronics, which aims to utilize the spin degree of freedom of electrons. Although early demonstrations of spin injection into STO-based 2DEG were hindered by spin-trap states in the insulator layer used to form the 2DEG, ferromagnetic resonance measurements have suggested a long spin diffusion length (>100 nm) up to room temperature [1]. However, direct evidence of spin-polarized transport in STO-based 2DEG has not yet been demonstrated, whereas highly crystalline perovskite transition-metal oxides are known to support ballistic spin-polarized transport. In La_{1-x}Sr_xMnO₃ (LSMO)-based systems, where Mn 3d orbitals contribute to electronic transport, a large spin-valve ratio of 140% and electron coherence in quantum-well structures have been demonstrated [2,3] despite the strong electron correlation of Mn 3d electrons.

Previous works [4,5] aimed at spin-polarized carrier injection from a ferromagnet source into an STO-based 2DEG. In their configurations, the 2DEG channel is at the interface between an insulator (LaAlO₃) and STO. In such structures, however, the insulator separates the ferromagnet from the 2DEG, resulting in a loss of spin polarization. A solution to this problem is to prepare a 2DEG via Ar irradiation that does not have an insulator layer [6].

In this report, we demonstrate spin-polarized carrier transport in a two-terminal spin-valve device consisting of ferromagnetic Fe electrodes deposited on an STO substrate by molecular beam epitaxy [Fig. 1(a)]. A 100-nm-long 2DEG channel was formed between the Fe electrodes by introducing oxygen vacancies through Ar irradiation etching. Clear spin-valve signals were observed at room temperature [Fig. 1(b)]. The electron mean free path is below 10 nm at temperatures above 100 K [Fig. 1(c)], indicating diffusive transport under these conditions. The persistence of the spin-valve signal suggests a spin lifetime on the order of nanoseconds [7]. At 50 K, however, a spin-valve signal with a magnetoresistance ratio of 1% was observed [Fig. 1(b) inset], while the mean free path increased to ~50 nm, enabling quasiballistic carrier transport.

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

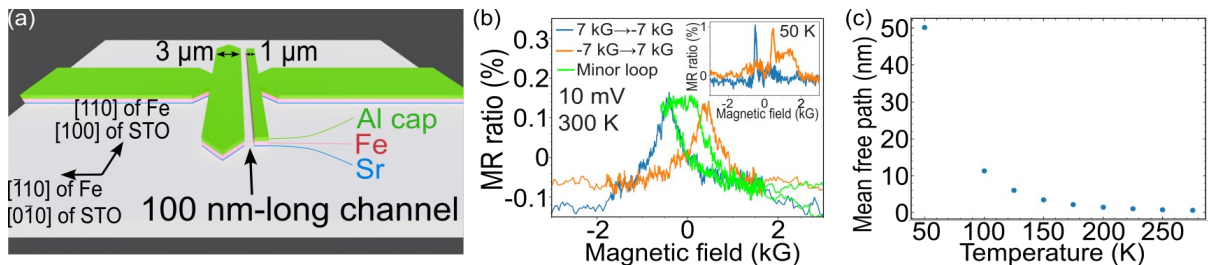


Figure 1 (a) Schematic illustration of the spin-valve device structure. The green/pink/blue and gray regions represent the Al/Fe/Sr layers and SrTiO₃ surface, respectively. (b) Spin-valve signal of the device measured at room temperature with an applied bias voltage of 10 mV. The inset shows the spin-valve signal at 50 K. (c) Electron mean free path in the 2DEG system prepared by the Ar irradiation in this study.

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Configurational-Entropy-Enabled Tunable Relaxor Transition and Dielectric Response Broadening in High-Entropy Ferroelectric Thin Films

Chun-Chung Hsu¹, Yu-Hsien Lee¹, Yi-Chun Chen², Yi-De Liou³, Ming-Hao Lee⁴, Ying-Hao Chu^{1*}

¹ *Department of Materials Science and Engineering, National Tsing Hua University, Taiwan*

² *Department of Physics, National Cheng Kung University, Taiwan*

³ *Department of Physics, National Changhua University of Education, Taiwan*

⁴ *Center for Condensed Matter Sciences, National Taiwan University, Taiwan*

Relaxor ferroelectrics exhibit slim polarization–electric field (P–E) loops, low coercive fields, reduced remanent polarization, and frequency-dependent dielectric responses associated with local structural disorder and dynamic polar nanoregions (PNRs). These characteristics make them attractive for dielectric, capacitor, and energy-storage applications. However, systematic tuning of relaxor transition behavior remains challenging because conventional relaxor systems, such as $\text{Pb}(\text{Mg,Nb})\text{O}_3\text{--PbTiO}_3$ (PMN-PT), are highly sensitive to composition and processing conditions, limiting their accessible design space for transition tuning.

In this work, configurational entropy is introduced to expand compositional flexibility and enable controllable relaxor behavior in ferroelectric thin films. A six-cation high-entropy perovskite system with configurational entropy approaching $1.75R$ is constructed by regulating the relative contribution of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ (PZT) and a multi-component B-site precursor, $\text{Pb}(\text{Mg,Nb,Ta,Ni})\text{O}_3$ (PMNTN), enabling investigation into how compositional complexity influences relaxor transition behavior. Because incorporation of multi-component B-site species alters the balance between ferroelectric ordering and relaxor behavior, systematic compositional variation provides a pathway to continuously tune the dielectric maximum temperature (T_m) and relaxor responses. Through this composition-driven strategy, T_m can be shifted toward the desired operating regime, enabling tunable relaxor responses under ambient conditions. Electrical characterization reveals systematic evolution from ferroelectric-like polarization behavior toward relaxor-like responses with composition and entropy variation. Increased configurational complexity is accompanied by enhanced local dipolar disorder and reduced ferroelectric coherence, consistent with the evolution toward finer polar nanoregions (PNRs). In addition to modulation of T_m , increased configurational entropy broadens the dielectric transition and reduces the temperature dependence of polarization response. Despite the compositional dependence of T_m , samples with higher configurational entropy exhibit improved polarization retention over temperature, suggesting that configurational entropy contributes not only to transition position through compositional variation but also to dielectric transition broadening and extension of the effective relaxor operating window.

These findings demonstrate that composition and configurational entropy jointly tune both the position and breadth of relaxor transitions, providing a practical strategy for designing dielectric and ferroelectric materials with tailored operating characteristics for next-generation electronic and energy-storage applications.

Potential realization of geometrical ferroelectricity in iron-based Grenier epitaxial thin films

Si Won Kim¹, Yong-Jun Kwon², Yongjin Shin³, Chan-Ho Yang² and Woo Seok Choi¹

¹ Department of Physics, Sungkyunkwan University, Suwon 16419, Republic of Korea

² Department of Physics, KAIST, Daejeon 34141, Republic of Korea

³ Department of Semiconductor Convergence Engineering, Dankook University, Yongin 16890, Republic of Korea

Grenier structure, a type of oxygen-vacancy-ordered perovskite, is characterized by an alternate stacking of octahedral bilayer and tetrahedral monolayer, i.e., an O-O-T polyhedral layer stacking (O: Octahedra, T: Tetrahedra) (Left panel of Figure 1) [1]. Recently, a stable ferroelectric ground state was theoretically predicted in Grenier oxides, based on density functional theory calculation [2]. This prediction points to geometrical ferroelectricity, where the polar state is primarily driven by the specific lattice distortion. The unique O-O-T polyhedral layer stacking enables tetrahedral distortion, such that the distortion leads to cation movements towards the same direction, along with accompanying octahedral distortion. Through this cooperative distortion of octahedral and tetrahedral layers, inversion symmetry breaking in an antiferromagnetic iron-based Grenier oxides having chemical formula of $R_{1/3}A_{2/3}\text{FeO}_{2.67}$ (R: Rare-earth elements, A: Alkaline earth metals) were suggested.

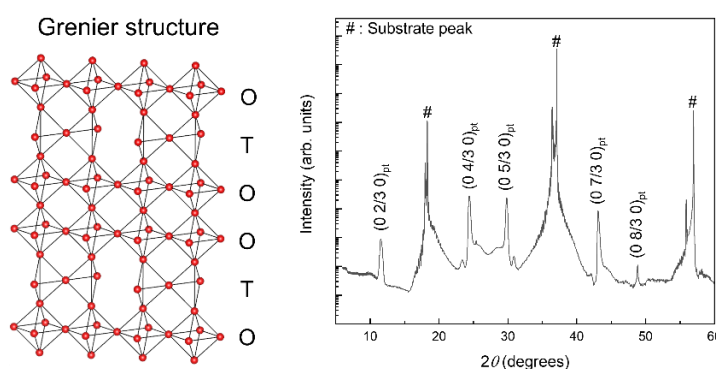


Figure 1: Schematic representation of Grenier structure and experimental XRD normal scan of $\text{Sm}_{1/3}\text{Sr}_{2/3}\text{FeO}_{2.67}$

Here, we propose observation of geometric ferroelectricity in epitaxial Grenier thin films. The films were synthesized by sub-unit-cell-scale precision modulation of brownmillerite $\text{SrFeO}_{2.5}$ and perovskite SmFeO_3 or NdFeO_3 using pulsed laser epitaxy. Synchrotron X-ray diffraction (XRD) measurements unambiguously show 1/3-order diffraction peaks which results from the unit cell tripling from the O-O-T stacking of the Grenier phase (Right panel of Figure 1). Insulating electronic structure with the optical band gap of ~ 2.1 eV of our film was characterized using spectroscopic ellipsometry. The observed O-O-T stacking led to a ferroelectricity, which is confirmed by piezoresponse force microscopy. Because the polar axis is perpendicular to the film normal, in-plane piezoresponse force microscopy was employed. This strategic synthesis of oxygen-vacancy-ordered structures opens new platform for tailoring inversion symmetry breaking in complex oxides through atomic-scale layer engineering.

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Ferroelectricity and magnetic order at room temperature and electronic states of bismuth iron oxyfluoride thin films

Akiko Kamigaito¹, Mizuho Sano¹, Kei Shigematsu², Satoshi Demura³, Hiroshi Kumigashira⁴, Tsukasa Katayama⁵, Yasushi Hirose⁶, Akira Chikamatsu¹

¹ *Department of Chemistry, Ochanomizu University*

² *Materials and Structures Laboratory, Institute of Integrated Research, Institute of Science Tokyo*

³ *Department of Physics, College of Science & Technology, Nihon University*

⁴ *Institute of Multidisciplinary Research for Advanced Materials, Tohoku University*

⁵ *Department of Applied Chemistry, Tokyo University of Science*

⁶ *Department of Chemistry, Tokyo Metropolitan University*

Iron oxyfluorides with large tetragonal polar distortions are expected to exhibit multiferroic properties that coexist with their ferroelectric and magnetic orders. In recent years, polycrystalline samples of Ba-doped bismuth iron oxyfluoride, $\text{Bi}_{1-x}\text{Ba}_x\text{FeO}_{3-x}\text{F}_x$ ($x = 0.2, 0.3$) have been reported to exhibit magnetic ordering [1]. First-principles calculations have also predicted that the composition with $x = 0.25$ possesses multiferroic properties [2]. However, oxyfluoride multiferroicity has not been experimentally confirmed in polycrystals, owing to their high leakage currents and the presence of impurity phases. In this study, we fabricated single-crystalline Ba-doped bismuth iron oxyfluoride thin films by combining pulsed laser deposition (PLD) with topochemical fluorination and investigated their room-temperature ferroelectric and magnetic properties.

The substitution of Ba at the Bi site in the precursor films was found to play an important role in the introduction of fluorine into the precursor by energy-dispersive X-ray spectroscopy. Figure 1 shows the polarization vs electric field (P - E) and the in-plane magnetization vs magnetic field (M - H) curves for the $\text{Bi}_{1-x}\text{Ba}_x\text{FeO}_{3-x}\text{F}_x$ ($x = 0.2, 0.3$) films. They exhibited ferroelectricity and a small magnetization at 300 K. Moreover, electronic structures due to the F 2p states were formed in the valence and conduction bands of the $\text{Bi}_{1-x}\text{Ba}_x\text{FeO}_{3-x}\text{F}_x$ ($x = 0.2, 0.3$) films. The findings demonstrate the potential of oxyfluoride compounds, thereby broadening the range of materials that can be used as room-temperature multiferroics.

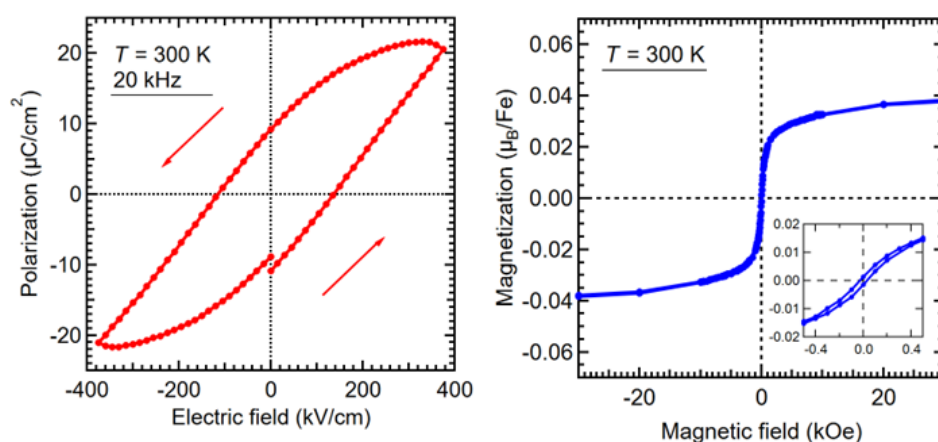


Figure 1: The $\text{Bi}_{1-x}\text{Ba}_x\text{FeO}_{3-x}\text{F}_x$ ($x = 0.2, 0.3$) films exhibited ferroelectricity and a small magnetization at 300 K.

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Engineering Interfacial Charge Transfer in SrNbO₃/SrTiO₃ heterostructures via SrZrO₃ barrier layers toward the development of transparent oxide transistors

Yuuki Masutake¹, Ryotaro Hayasaka¹, Seitaro Inoue¹, Daisuke Shiga^{1,2}, Kenichi Ozawa², and Hiroshi Kumigashira^{1,2}

¹ *Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, Sendai 980–8577, Japan*

² *Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba 305–0801, Japan*

Recently, the conductive oxide SrNbO₃ (SNO) has attracted significant attention as a promising transparent electrode material due to its high transparency extending into the UV region [1] and exceptionally high carrier mobility [2]. While heterojunctions between SNO and *n*-type transparent semiconductor SrTiO₃ (STO) are expected to enable a high-performance transparent Schottky junctions, a critical issue arises at the interface: inherent charge transfer from SNO to STO [3]. This unwanted charge transfer induces a parasitic metallic state on the STO side, which significantly degrades device performance by creating a leakage path. Thus, it is essential to establish a method to suppress this charge transfer for practical device applications.

In this study, we investigated the effectiveness of inserting a transparent insulating SrZrO₃ (SZO) barrier layer to suppress this interfacial charge transfer. Using laser molecular beam epitaxy with atomic-scale digital control, we fabricated SNO/SZO/STO heterostructures and investigated the change in the inherent charge transfer between the SNO and STO by inserting a SZO barrier layer with controlled layer thickness. To probe the buried interfacial electronic structure for the STO side, we employed *in-situ* Ti 2*p*–3*d* resonant photoemission spectroscopy. By utilizing the elemental selectivity of Ti 2*p*–3*d* resonant photoemission spectroscopy for the Ti 3*d* state near the Fermi level (E_F), we directly visualize the suppression of charge transfer as a function of barrier thickness. We found that the Ti 3*d* spectral weight near E_F decays exponentially with increasing SZO thickness, yielding a characteristic decay length (λ) of 1.0–1.2 nm. Notably, this value is significantly longer than the 0.3 nm predicted by a simple electron tunneling model, suggesting that the chemical bonding between transition metals and oxygen plays an important role in the charge propagation. Our findings provide guidelines for designing and controlling functionalities in oxide nanostructures.

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Flexible In₂O₃ Thin-Film Transistors with High-Mobility ($\sim 75 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$)

Hyeonjun Kong¹, Ying-Hao Chu², Yasutaka Matsuo³, and Hiromichi Ohta⁴

¹ *Graduate School of Information Science and Technology, Hokkaido Univ.*

² *Department of Materials Science and Engineering, National Tsing Hua Univ.*

³ *Research Institute for Electronic Science, Hokkaido Univ.*

Oxide semiconductor thin-film transistors (TFTs) have become key switching devices in modern flat-panel display backplanes. Although currently applied TFTs show mobility of $\sim 10 \text{ cm}^2/\text{Vs}$, in the near future, TFTs will require higher mobility $> 70 \text{ cm}^2/\text{Vs}$ because higher drive current and faster pixel switching are required in advanced flat panel displays. Hydrogen-incorporated In₂O₃ is a particularly promising channel material because of its high mobility ($\mu_{\text{FE}} \sim 140 \text{ cm}^2/\text{Vs}$) [1,2]. Although it was difficult to introduce hydrogen gas to the film, recently, we fabricated In₂O₃ TFTs by PLD using In(OH)₃ ceramic targets without introducing hydrogen gas [3]. The next challenge is to transfer this high-mobility In₂O₃ TFT technology from rigid substrates to mechanically flexible substrates while minimizing degradation of the channel quality required for high μ_{FE} . This is difficult for many conventional flexible substrates, whose thermal processing limits are typically around 220 °C and therefore restrict the crystallization process needed for high-quality In₂O₃ growth. In this study, we demonstrate high-mobility In₂O₃ TFTs fabricated on flexible mica substrates using In(OH)₃ targets. Mica substrates offer a unique combination of mechanical flexibility, an atomically flat cleavage surface, and high thermal stability [4]. In particular, the atomically flat surface can suppress roughness-induced carrier scattering, while the high thermal stability allows the thermal processing required to promote large-grain In₂O₃ growth. The fabricated TFTs exhibit a high μ_{FE} of $\sim 75 \text{ cm}^2/\text{Vs}$, confirming that mica can serve as an effective platform for high-mobility flexible oxide TFTs. Furthermore, cyclic bending tests up to 1000 cycles were also conducted to evaluate the mechanical robustness of the In₂O₃ films and to examine how bending-induced film degradation affects the electrical performance of flexible TFTs.

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All-Solid-State Redox Transistor Using VO₂ Thin Film and Li⁺ Solid Electrolyte

Tohru Higuchi and Taisei Yamada

Department of Applied Physics, Tokyo University of Science, Tokyo 125-8585, Japan

In recent years, Mott transistors have attracted attention as low-power memory devices, and there are expectations for the realization of new devices in which resistance can be controlled by applying voltage to strongly correlated electron materials. Among these, VO₂ exhibits a metal-insulator transition (MIT) with a resistance change of more than three orders of magnitude around 340 K, and there is a possibility that its characteristics can be controlled by modulating the electron carrier concentration. In this study, we designed an all-solid-state transistor using a VO₂ thin film and a Li⁺ solid electrolyte, Li₃PO₄(LPO)/LiCoO₂(LCO), and attempted to modulate the electrical properties of the VO₂ thin film by oxidation-reduction reactions associated with the Li⁺ insertion and desertion.

An all-solid-state transistor as shown in Fig. 1 was fabricated by depositing a VO₂ thin film, which serves as the channel layer, on an Al₂O₃(0001) substrate using RF magnetron sputtering, a LPO thin film as the Lithium ion conduction layer, and LCO and Pt as lithium ion supply layers for the electrodes. V-metal and LPO/LCO ceramic targets were used as targets. The crystal structure of the fabricated VO₂ thin film was evaluated by X-ray diffraction, and the temperature dependence of its electrical resistance was measured by a DC two-terminal method. The ionic conductivity of the LPO and LCO thin films was evaluated by AC impedance spectroscopy. The I-V characteristics of the all-solid-state transistor were evaluated by gate voltage (V_G) sweep measurement, determining the drain current I_D and gate current I_G.

The prepared VO₂ single layer exhibited a four-order-of-magnitude MIT based on the temperature dependence of its resistance. The impedance measurements confirmed that the LPO and LCO thin films showed clear Li⁺ conduction. As shown in Fig. 2, the fabricated all-solid-state transistor had a relatively high I_D (~0.5 μA) at the initial state (V_G=0V), suggesting that a Li⁺ insertion reaction into VO₂ ($\text{VO}_2 + x\text{Li}^+ + xe^- \rightarrow \text{Li}_x\text{VO}_2$) proceeded during the film deposition process, forming Li_xVO₂. In process ①, where negative V_G was applied, the I_D increased by 496.8% as Li⁺ in Li_xVO₂ moved to the LCO side as V_G decreased. In process ②, where V_G was applied from negative to positive, the I_D decreased by 693.0% as Li⁺ supplied from the LCO side was inserted into VO₂ as the voltage increased. These results suggest that the reversible insertion and removal of Li⁺ by applying a gate voltage affects the electrical conductivity characteristics of VO₂, indicating that this is an all-solid-state redox transistor capable of modulating the VO₂ channel resistance.

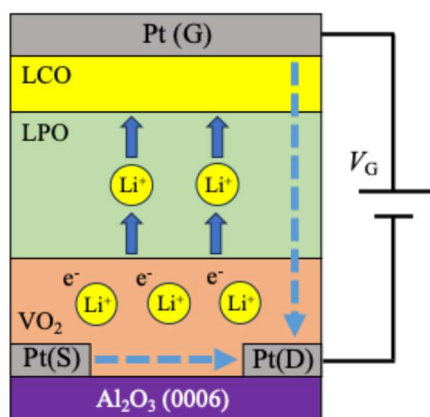


Fig.1 Schematic diagram of all-solid-state transistor

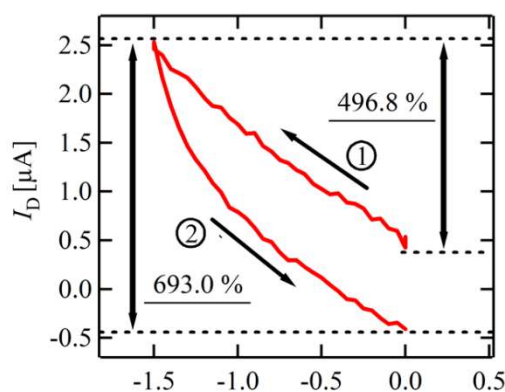


Fig. 2 I_D-V_G characteristics of all-solid-state transistors

Solution-Processed All-Metal-Oxide Permeable-Base Transistors for High-Current-Density Vertical Oxide Electronics

Ertürk E. Yildirim¹, Mike Hambsch¹, Markus Löffler², Shivam Singh³, Yana Vaynzof³, Bernd Rellinghaus², Stefan C.B. Mannsfeld¹

¹*Faculty of Electrical and Computer Engineering and Center for Advancing Electronics Dresden (cfaed), TUD Dresden University of Technology, Dresden, 01062, Germany.*

²*Dresden Center for Nanoanalysis (DCN), Center for Advancing Electronics Dresden (cfaed), TUD Dresden University of Technology, Dresden, 01062, Germany.*

³*Dresden Integrated Center for Applied Physics and Photonic Materials (IAPP), TUD Dresden University of Technology, Dresden, 01062, Germany.*

Indium-gallium-zinc oxide (IGZO) has emerged as a leading amorphous oxide semiconductor for thin-film transistors (TFTs), owing to its combination of low-temperature processability, mechanical flexibility, optical transparency and high carrier mobility. These properties make IGZO highly attractive for active-matrix OLED backplanes, where pixel-driving transistors must provide large current densities at low operating voltages while occupying minimal lateral area. However, planar IGZO TFTs face scaling limitations that restrict the achievable drive current and switching performance, motivating the exploration of vertical architectures, which often require complex fabrication. Here, we report the first all-metal-oxide-semiconductor permeable-base transistor (MOPBT) using an environmentally friendly, water-based solution process, introducing a new vertical three-terminal architecture for oxide electronics. By employing a porous base electrode to modulate vertical charge transport, the device achieves current densities up to 1 A cm^{-2} , high current gain β of 24000, and efficient low-voltage switching within a simple layer-by-layer fabrication scheme. The unique properties of IGZO demonstrated in this work also make it highly suitable for vertical integration schemes for OLEDs and other emerging electronic applications.

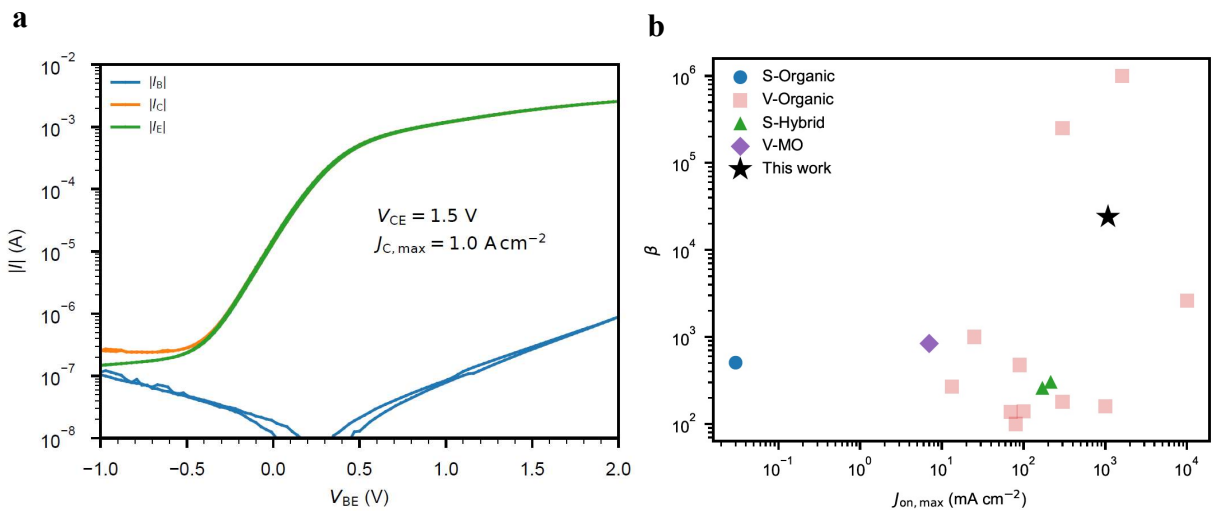


Figure 1: a) Transfer characteristics of a MOPBT in common-emitter configuration at $V_{CE} = 1.5 \text{ V}$, exhibiting a maximum current density of 1.0 A cm^{-2} and an on/off ratio of 10^4 . b) Literature comparison of permeable-base transistors classified by semiconductor and processing route: solution-processed organic (S-Organic), vacuum-processed organic (V-Organic), solution-processed hybrid organic/metal-oxide (S-Hybrid), and vacuum-processed metal-oxide (V-MO) devices. Current gain (β) is plotted against maximum on-state current density ($J_{on,max}$).

Human-Interactive Reservoir Computing using an Oxide Ionic Drift-Diffusion Transistor

H. Inoue¹, H. Tamura^{2,3}, A. Kitoh¹, X. Chen⁴, Z. Byambadorj⁴,
T. Yajima⁵, Y. Hotta⁶, T. Iizuka⁴, G. Tanaka^{3,7}, and I. H. Inoue¹

¹ *National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan*

² *Graduate School of Informatics, Department of Core Informatics, Osaka Metropolitan University,
Osaka, Japan*

³ *International Research Center for Neurointelligence (IRCN), The University of Tokyo, Tokyo,
Japan*

⁴ *Systems Design Laboratory, School of Engineering, The University of Tokyo, Tokyo, Japan*

⁵ *Graduate School of Information Science and Electrical Engineering, Kyushu University, Fukuoka,
Japan*

⁶ *Department of Engineering, University of Hyogo, Hyogo, Japan*

⁷ *Department of Computer Science, Nagoya Institute of Technology, Nagoya, Japan*

Oxide materials offer unique opportunities for electronic devices beyond the limits of conventional silicon technology. In particular, their ionic character enables the coexistence of fast electronic conduction and controllable ionic transport within a single material. Because ionic motion is much slower than electronic motion and can retain its state nonvolatily, it provides a natural basis for dynamic and memory functions governed by an external electric field. Here, we explore such ionic dynamics for energy-efficient information processing.

Reservoir computing is a neural-network-inspired framework, motivated in part by biological neural networks, that exploits the intrinsic dynamics of a physical system to process temporal information [1]. For such computation, a device should exhibit a history-dependent response with fading memory. Leaky integration, a characteristic dynamical feature of biological neurons, is one representative example. We have developed an ionic drift-diffusion transistor based on the oxide semiconductor SrTiO₃ that reproduces this behavior on biologically relevant timescales of about 1 s, making it promising for human-interactive sensing and processing [2].

Building on this leaky integration, we implemented leaky-integrate-and-fire functionality by combining the transistor with simple circuit elements, thereby extending the device operation from fading memory to active spike generation. We further constructed a small reservoir network based on these elements. The resulting circuit exhibits nonmonotonic spiking responses, indicating the nonlinear transient dynamics required for reservoir computing [3]. We will also present simulation results showing how such devices can perform meaningful temporal information processing. These findings highlight the potential of oxide ionic devices as a platform for ultralow-power edge computing in isolated or energy-harvesting environments.

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First Demonstration of Suboxide Molecular-beam epitaxy Grown p-Type SnO TFT

Muhaimin Mareum Khan¹, Seungmin Lee¹, Anna Sunah Park¹, Joseph E Dill¹, Madhav Ramesh¹, Bennett J Cromer¹, Debdeep Jena¹, Darrell G Schlom¹, and Huili Grace Xing¹

¹ Cornell University, Ithaca, NY 14853, USA

The development of high-performance p-type oxide semiconductors remains a critical bottleneck for realizing all-oxide complementary metal-oxide-semiconductor (CMOS) technology, particularly for back end of line (BEOL)-compatible monolithic 3D integration. Among candidate materials, α -SnO is particularly promising due to its hole mobility reported as high as $21 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ [1] and low-temperature processing window below 400°C . However, fabricating p-type SnO thin-film transistors (TFTs) with reliable electrostatic control remains challenging because SnO surface is highly sensitive to standard processing solutions and thermal treatments above 350°C .

In this work, we report the first realization of α -SnO TFTs grown by suboxide molecular-beam epitaxy (MBE). Our initial experiments showed that the SnO sheet carrier density can increase by up to five times after device processing, making the channel difficult to pinch off and causing high off-state current. We are currently investigating the origin of this process-induced surface carrier increase. However, we found that tetramethylammonium hydroxide (TMAH), a standard photoresist developer, increases the SnO surface charge density and degrades electrostatic control. Similar chemical sensitivity was reported by Wang et al., who observed that TMAH attacks ZnO films [2]. Bae et al. [3] showed that an ALD Al_2O_3 capping layer improves the performance and stability of p-channel SnO TFTs, attributing this to reduced oxygen vacancy induced hole trap states due to hydrogen diffusion from Al_2O_3 . Inspired by this work, we introduced a 200°C thermal ALD Al_2O_3 passivation layer to protect the SnO channel from degradation during processing.

Using this surface-protection strategy, SnO TFTs with an $18 \mu\text{m}$ gate length, channel thickness 10.3 nm and Hall mobility $4 \text{ cm}^2/\text{Vs}$ exhibit an on/off current ratio of 1.62×10^2 , a maximum current density of $\sim 0.74 \mu\text{A}/\mu\text{m}$, and gate leakage below $10^{-8} \mu\text{A}/\mu\text{m}$, as shown in Fig. 1(a,b,c). Because the optical bandgap of SnO is approximately 2.7 eV , all electrical measurements were performed in the dark to minimize photoinduced effects. The gate leakage is approximately six orders of magnitude lower than the source-drain current, confirming that the measured transistor characteristics are not gate-leakage-limited. We are currently investigating the effect of ALD Al_2O_3 on the structural, electrical transport, and Hall properties of SnO.

In parallel with channel processing and gate-dielectric optimization, we performed a systematic contact study to minimize contact resistance, R_c , which is essential for scaling to short-channel ($<1 \mu\text{m}$) devices. Leveraging controllable K doping, first time demonstrated by the Schlom group at Cornell University [4], we studied high-work-function Ni and Pt contacts to potassium-doped α -SnO channels with acceptor concentrations from 5×10^{17} to $5 \times 10^{19} \text{ cm}^{-3}$ (Fig. 1d). Fully BEOL-compatible Pt contacts annealed in air at 300°C for 10 min yielded the best contact performance, achieving a low R_c of $1.9 \text{ k}\Omega \cdot \mu\text{m}$ and a low ρ_c of $3.9 \times 10^{-6} \Omega \cdot \text{cm}^2$. These results establish MBE-grown α -SnO as a promising p-type oxide semiconductor for BEOL-compatible oxide electronics. Ongoing work focuses on further improving the on/off ratios and device performance through channel thickness and doping optimization to enable full carrier depletion, as well as fundamental studies of the chemical reaction mechanisms between the SnO surface and TMAH and Al_2O_3 .

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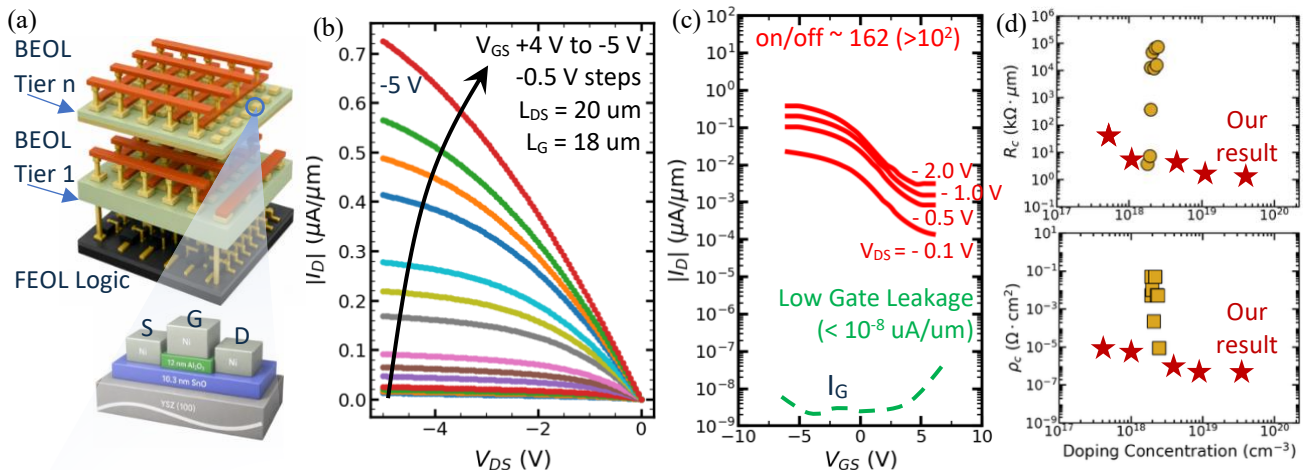


Fig. 1 (a) Schematic of the BEOL-compatible p-type oxide SnO TFT consisting of a 10.3 nm SnO channel, 12 nm Al_2O_3 dielectric/passivation layer, and Ni contacts used for source, drain, and gate electrodes. (b) Output family curve showing a maximum current density of $\sim 0.74 \mu\text{A}/\mu\text{m}$ at $V_{GS} = -5 \text{ V}$. (c) Transfer characteristics measured at $V_{DS} = -0.1 \text{ V}$, showing an on/off current ratio ~ 162 ($>10^2$) and very low gate leakage current $\sim 10^{-8} \mu\text{A}/\mu\text{m}$. (d) Benchmarking our R_c and ρ_c against reported values for SnO contacts, where prior studies are very limited, unsystematic, and under-documented, and were performed only on UID SnO.

Harnessing Surface-Dominant Proton Transport for Reservoir Computing in VO₂ Two-Terminal Devices

Satoshi Hamasuna¹, Takeaki Yajima¹

¹ Faculty of Information Science and Electrical Engineering, Kyushu University, Japan

Proton (H⁺) doping in complex oxides can modulate various physical properties. H⁺ can be incorporated uniformly and reversibly via voltage application, enabling neuromorphic functionalities with memristive and nonlinear characteristics. In canonical mixed conductors such as WO₃, H⁺ doping induces metallization, which screens the applied electric field and limits reversible voltage control. As a result, three-terminal device geometries have been widely adopted [1]. In contrast, rare-earth nickelates and vanadium dioxide (VO₂) become more insulating upon H⁺ doping [2], facilitating voltage control even in two-terminal devices. However, the H⁺ transport in two-terminal mixed conductors remains poorly understood. This study reveals that surface-dominant H⁺ transport in Pt/VO₂/Pt two-terminal devices can serve not only as a mechanism for reversible ion control but also as a computational resource for reservoir computing.

Measurements on Pt/VO₂/Pt two-terminal devices (Fig. 1a) revealed that efficient H⁺ doping requires the high-mobility *c_R* axis to be aligned out of plane rather than along the VO₂ channel direction (Fig. 1b). These results indicate the surface transport of H⁺ followed by incorporation into VO₂ along the *c_R* axis. Because the applied voltage drops primarily across the insulating H⁺-doped region, fully reversible resistance modulation was achieved (Fig. 1c). The relaxation behavior naturally provides short-term memory for reservoir computing. A compact five-device reservoir (Fig. 1d) was applied to blood-glucose time-series denoising, and successfully learned a first-order low pass filter despite device nonlinearity and variability (Fig. 1e). Owing to its capacitor-free design, the proposed filter is highly scalable compared with conventional RC filters.

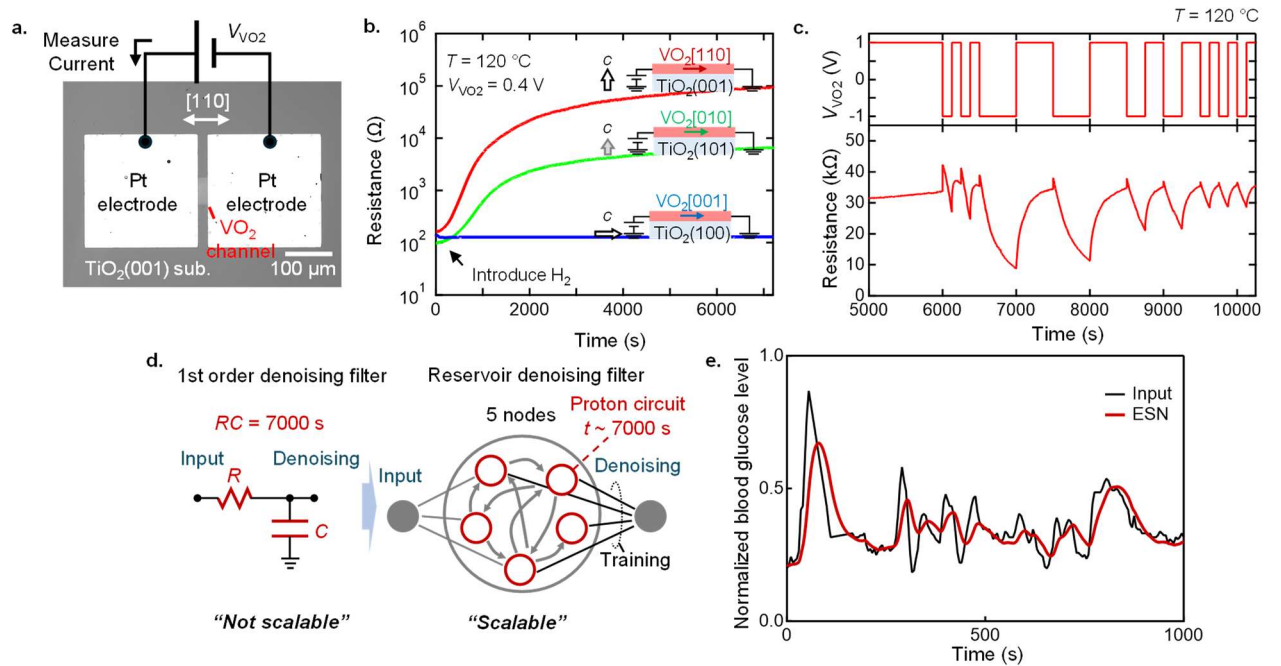


Figure 1: **a**, Optical scope image of the two-terminal device. **b**, Proton doping characteristics (Resistance vs. time) as a function of crystal orientation at $T = 120\text{ }^{\circ}\text{C}$. **c**, Demonstration of voltage-controlled resistance modulation. **d**, LPF architectures: RC circuit (not-scalable) and Pt/VO₂/Pt reservoir (scalable). **e**, Blood-glucose time-series denoising.

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Low-Temperature RF Switching Device Using the Metal-Insulator Transition of W-Doped VO₂ Thin Films

Harutaka Koyama [1,2], Hiroshi Oike [2],

Keisuke Shibuya [3], Takanobu Watanabe [1], Yusuke Kozuka [1,2,4]

¹ Faculty of Science and Engineering, Waseda University, Shinjuku, Tokyo, Japan

² Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), Tsukuba, Ibaraki, Japan

³ National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki, Japan

⁴ WPI Advanced Institute for Materials Research, Tohoku University, Sendai, Miyagi, Japan

In quantum computers, as the number of qubits increases, a large number of coaxial cables are used in the cryostat. These numerous coaxial cables not only contribute to the heat load but also limit the available space within the cryostat. To overcome this limitation, one possible way is to use RF switches in the cryostat. However, most commercial RF switches cause nontrivial heating or require large operation voltages [1,2]. These properties are not suitable for application at low temperatures, where the cooling power is limited.

In this study, we developed RF switching devices that operate at cryogenic temperatures using tungsten-doped VO₂ (W:VO₂). Undoped VO₂ is known to undergo a metal-insulator transition near room temperature [3]. By substituting V with W, the metal-insulator transition temperature is lowered as a result of carrier doping [4], enabling RF switching at low temperatures. Here, we have fabricated W:VO₂ thin films on TiO₂ (001) substrates using pulsed laser deposition. The W:VO₂ thin film was then patterned with reactive ion etching, and Ti/Au layers were deposited as shown in Fig. 1 to form an RF waveguide with W:VO₂ implemented as a switching element.

The temperature dependence of the RF transmission characteristics is shown in Fig. 2. This figure shows that RF transmission (S_{21}) is significantly suppressed below 150 K as a result of the metal-insulator transition, demonstrating that the waveguide works as a temperature-driven RF switching device. This study represents the first demonstration of a low-temperature RF switching device using W:VO₂ for its potential application as an RF switching device in a cryostat.

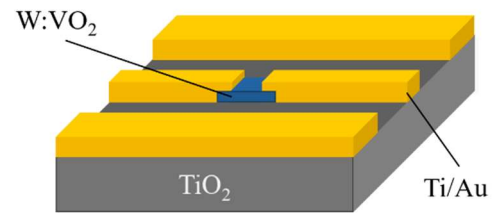


Figure 1: Schematic diagram of an RF switching device using W:VO₂.

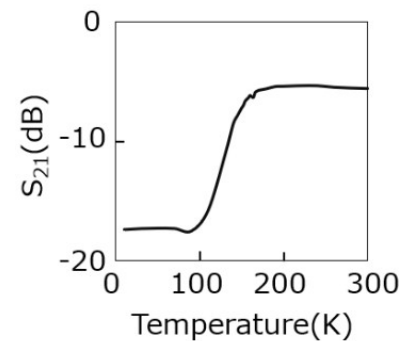


Figure 2: Temperature dependence of S_{21} (RF frequency = 1GHz)

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Amorphous, Highly Conductive $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ for Area-Dependent Resistive Switching Devices

M. Buczek¹, I. T. Dossayev¹, C. Wittberg¹, Y.-P. Liu¹, K. Goß¹, D. N. Mueller^{1,2}, Z. Moos¹, Z. Liu¹, K. Bittkau³, M. H. Sultani¹, S. Menzel¹, S. Hoffmann-Eifert¹, R. Dittmann¹

¹ Peter-Grünberg-Institute 7, Forschungszentrum Jülich, Germany

² Peter-Grünberg-Institute 6, Forschungszentrum Jülich, Germany

³ Institute of Energy Materials and Devices, Forschungszentrum Jülich, Germany

Memristive $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (PCMO) heterostructures exhibit area-dependent resistive switching via a valence change mechanism, making them promising for neuromorphic architectures [1]. A major challenge in PCMO-based memory is higher-dimensional lattice defects that affect oxygen-vacancy migration and concentration. This study mitigates these defects using highly conductive amorphous PCMO fabricated via a CMOS back-end-of-line-compatible [2] process and compares it with low-conductive amorphous and polycrystalline PCMO. The resistance differences are attributed to changes in electronic mobility, based on the analysis of short- and long-range order, Mn–O hybridization, and Mn valence state. AlO_x /PCMO devices showed the highest ON/OFF ratio compared to low-conductive amorphous and polycrystalline PCMO, because the field-accelerated oxygen vacancy movement switches the mechanism from Poole–Frenkel emission in the LRS to trap-assisted tunneling in the HRS. The mechanism change was identified by systematically analyzing the I-V asymmetry, device band diagrams for different PCMO types, and shape changes in the I-V curve fits. The band diagrams were calculated from the measured band gaps and work functions of the different PCMO types. Analysis of the electric field distribution in the devices showed a clear correlation between the pre-switching field strength in AlO_x and the resulting ON/OFF ratio.

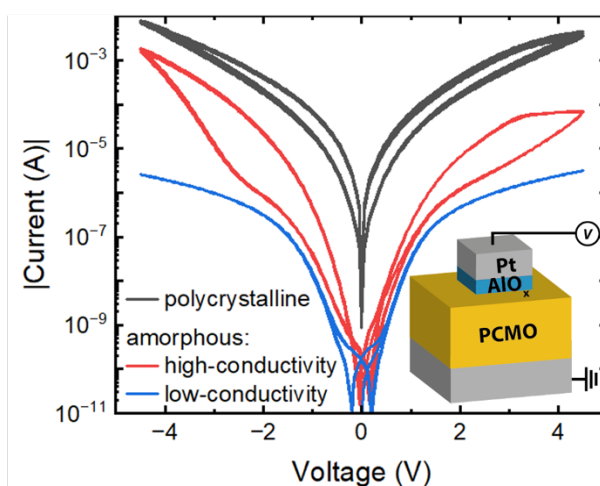


Figure 1: Highest on/off ratio for quasi-amorphous PCMO/ AlO_x devices compared to crystalline or amorphous PCMO

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Observation of Resistive Switching in NbO₂ Thin Films and Analysis Using a Two-State Model

Maya Umezaki¹, Hao-Bo Li^{1,2}, Hidekazu Tanaka^{1,2}

¹ *The University of Osaka, SANKEN*

² *Spintronics Research Network Division, OTRI*

Transition-metal oxides exhibiting a metal–insulator transition (MIT) have attracted considerable attention as promising materials for beyond-CMOS devices [1]. Among them, NbO₂ has a high transition temperature ($T_{MIT} \approx 807$ °C) and is expected to serve as a thermally stable switching material for high-temperature device applications. For practical device applications, however, it is essential to clarify the conditions under which switching occurs in thin films and to identify its underlying physical origin.

In this study, NbO₂ thin films were grown on Al₂O₃(0001) substrates by pulsed laser deposition, and their current–voltage characteristics were investigated. At room temperature, the devices exhibited nearly ohmic current–voltage behavior. In contrast, when the external heating temperature was increased to approximately 160 °C, threshold-like switching behavior accompanied by a nonlinear increase in current and pronounced hysteresis was observed. Furthermore, the devices returned to their initial high-resistance state after the applied voltage was removed, indicating volatile switching behavior.

Possible mechanisms for this switching behavior include a Schottky barrier formed at the metal electrode–semiconductor interface, ReRAM-type resistance switching caused by the migration of oxygen ions or oxygen vacancies, and a local MIT induced by Joule heating associated with current concentration. However, the observed volatile hysteretic behavior is difficult to explain solely by a simple Schottky-barrier mechanism or a conventional ion-migration-based mechanism. Therefore, the I–V characteristics were analyzed using a two-state model assuming the coexistence of insulating and metallic phases in NbO₂. The results suggest that, when the resistance ratio between the two phases is sufficiently large, current concentration promotes the formation of a local precursor state of the MIT, resulting in threshold switching at a relatively low external temperature.

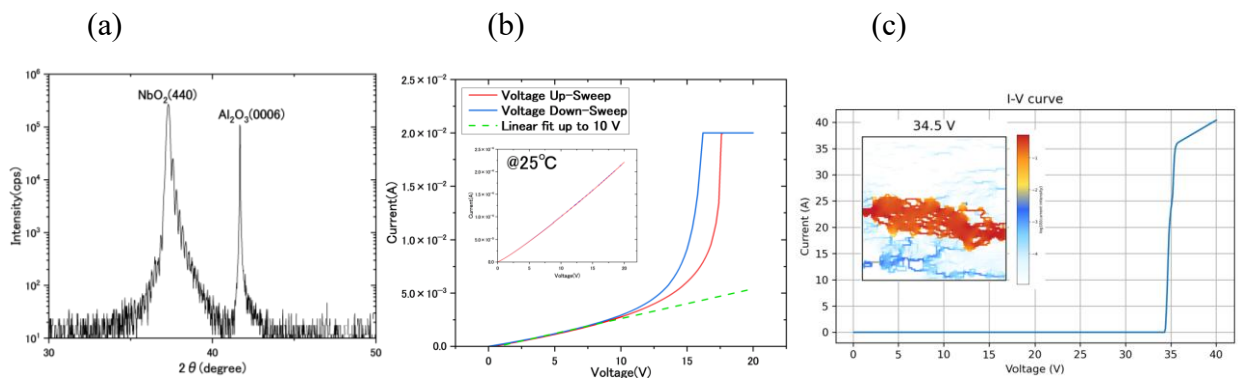


Fig. 1. (a) Typical X-ray diffraction pattern of a NbO₂ thin film on Al₂O₃(0001). (b) Current–voltage characteristics measured at 160 °C, showing threshold switching and hysteresis around 17 V. Inset: I–V characteristics at 25 °C. (c) Simulated I–V characteristics and current-density distributions for ($R_I/R_M = 10^5$), showing Joule-heating-induced threshold switching.

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[1] IRDS™ 2023: Beyond CMOS and Emerging Materials Integration

Tailoring a Novel $\text{Li}_{1.25}\text{Cr}_{0.25}\text{Ti}_{1.5}\text{O}_4$ Anode for Thin Film Microbatteries

E. Quintero¹, G. Koster¹, M. Huijben¹

¹ MESA+ Institute, University of Twente, 7500 AE Enschede, The Netherlands

Lithium-ion batteries offer a distinctive combination of high energy and power densities, which has established them as the leading technology for portable electronics. Despite their success, technical constraints associated with the use of liquid electrolytes and cell design make their integration into smaller devices challenging. To address this limitation, the novel spinel oxide $\text{Li}_{1.25}\text{Cr}_{0.25}\text{Ti}_{1.5}\text{O}_4$ (LCTO) is here proposed as an alternative anode material for solid-state Li-ion microbatteries.

In this work, highly crystalline LCTO thin films were deposited for the first time on top of Nb-doped SrTiO_3 (111) substrates by pulsed laser deposition (PLD). The films demonstrated a flat, smooth surface over a wide range of deposition temperatures with an out-of-plane crystal orientation parallel to the [111] direction. The structural properties of the spinel films were assessed by means of atomic force microscopy (AFM) and a comprehensive X-ray diffraction (XRD) analysis, encompassing X-ray reflectivity (XRR), symmetrical scans, rocking curves, and reciprocal space mapping (RSM). In addition, X-ray photoelectron spectroscopy (XPS) was used to probe the near-surface elemental composition and oxidation states, confirming the presence of Ti^{4+} and Cr^{3+} in the film as-deposited. Low-polarization flat potential plateaus centered around 1.55 V vs Li^+/Li in charge/discharge curves suggest that the LCTO redox mechanism proceeds through a two-phase reaction enabled by the $\text{Ti}^{3+}/\text{Ti}^{4+}$ redox pair. The same mechanism is observed in the state-of-the-art $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (LTO) anode material. Notably, the LCTO plateau discharge capacity of 160 mAh/g significantly surpassed the capacity previously reported for similar LTO thin films (120 mAh/g) [1]. More importantly, the total reversible capacity was enhanced up to roughly 230 mAh/g at C/8 by possibly storing extra Li capacity at the surface. These values are consistent with those documented in the literature for modified LTO formulations [2]. Moreover, the films demonstrated outstanding cycling stability as evidenced by the capacity recovery to 230 mAh/g for 1C after charging and discharging at 20C rate. Future work will focus on elucidating the excess lithium storage mechanisms, as well as evaluating the effect of Cr in the electrical and ionic conductivity of LCTO.

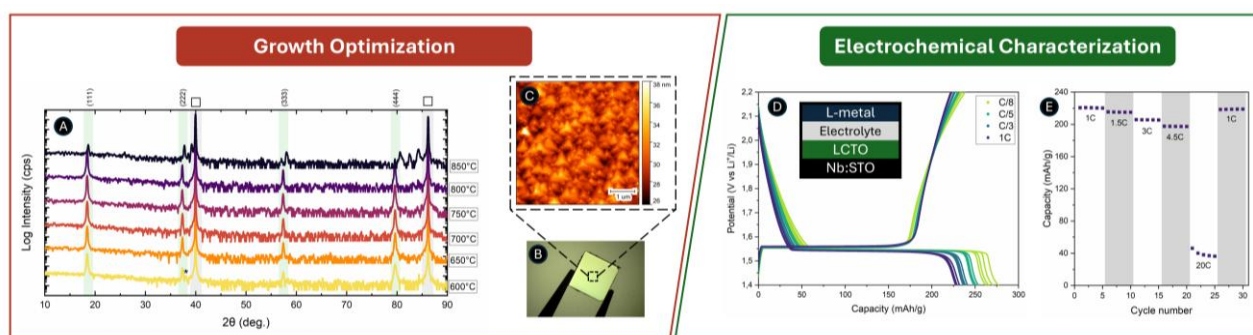


Figure 1: Schematic representation of XRD (A), optical microscope (B), and AFM (C) data recorded for the growth optimization of $\text{Li}_{1.25}\text{Cr}_{0.25}\text{Ti}_{1.5}\text{O}_4$ (LCTO) thin films. (D) and (E) show, respectively, the charge/discharge plot and reversible capacity measured in a half-cell for an LCTO film deposited at 750°C.

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Interfacial Engineering for Rapid Charging 5V-Class Thin-Film All-Solid-State Li Batteries

Kazunori Nishio¹, Asuka Oi², Kurei Edamura², Yuki Takeda², Shigeru Kobayashi¹,
Ryo Nakayama¹, Ryota Shimizu³, and Taro Hitosugi^{1,2}

¹ The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

² Institute of Science Tokyo, 2-12-1 Meguro-ku, Ookayama, Tokyo 152-8550, Japan

³ Institute for Molecular Science, 38 Nishigo-Naka Myodaiji, Okazaki, Aichi 444-8585, Japan

Rapidly chargeable all-solid-state batteries operating at high current densities are essential for next-generation high-power energy-storage systems. However, the electrolyte–electrode interfacial resistance in 5 V-class all-solid-state batteries remains a critical issue, particularly under high-current-density operation (14 mA cm^{-2} at 70°C). [1] In this study, we quantitatively investigate rapid charge–discharge behavior in thin-film all-solid-state Li batteries consisting of epitaxial $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) positive electrodes, Li_3PS_4 solid electrolyte, and Li metal negative electrodes fabricated entirely via non-air-exposure processes. [2] The thin-film platform enables quantitative evaluation of interfacial resistance using atomically well-defined interfaces.

Direct contact between Li_3PS_4 and LNMO resulted in extremely high interfacial resistance and prevented battery operation. Introducing a Li_3PO_4 buffer layer at the Li_3PS_4 –LNMO interface (Fig. 1a) drastically reduced the interfacial resistance by approximately 50,000-fold and enabled normal battery operation. However, rapid charge–discharge operation remained unstable at 30 mA cm^{-2} . In contrast, the stability during high-current-density cycling originating from the Li– Li_3PS_4 interface was improved by introducing 100 nm-thick Li_3PO_4 (Fig. 1c) or ultrathin Pt (Fig. 1d) buffer layers at the negative-electrode side. As a result, stable charge–discharge operation at a current density of 30 mA cm^{-2} at room temperature was achieved in a 5 V-class thin-film all-solid-state battery.

These results demonstrate the importance of quantitative interface engineering at both positive- and negative-electrode interfaces for realizing rapidly chargeable high-voltage all-solid-state batteries.

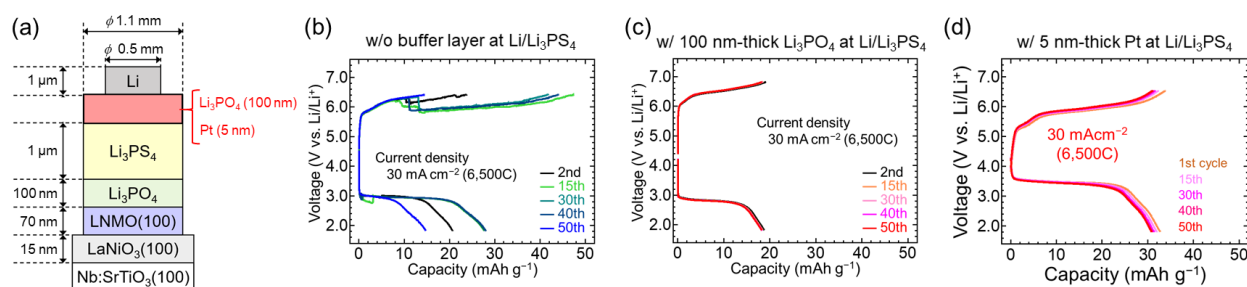


Figure 1 (a) Cross-sectional schematic of the thin-film battery. Charge–discharge curves at 30 mA cm^{-2} of batteries (b) without a buffer layer and with (c) a 100 nm-thick Li_3PO_4 buffer layer and (d) a 5 nm-thick Pt buffer layer at the Li– Li_3PS_4 interface.

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A Novel Transparent n-Type Semiconductor β -TeO₃

Kota Hanzawa¹, Tomotaka Fukunaga¹, Tianwei Wang^{1,2},

Fumiyasu Oba^{1,2,3}, and Hidenori Hiramatsu^{1,3}

¹ *Materials and Structures Laboratory, Institute of Integrated Research, Institute of Science Tokyo, Kanagawa, Japan*

² *Kanagawa Institute of Industrial Science and Technology (KISTEC), Kanagawa, Japan*

³ *MDX Research Center for Element Strategy, Institute of Integrated Research, Institute of Science Tokyo, Kanagawa, Japan*

Transparent semiconductors are key materials as carrier injection/transport layers for practical optoelectronic devices such as light-emitting diodes and photovoltaic cells. For hole injection, although Sn-doped In₂O₃ (ITO) and MoO_x have conventionally been employed, these materials face bottlenecks: an insufficient work function for ITO ($\phi = 4.7\text{--}5.6$ eV) and low transparency for MoO_x. Therefore, novel transparent semiconductors with a high ϕ are in high demand. In MoO_x, a high ϕ reaching 6.6 eV is predominantly attributed to the d⁰ electronic structure of Mo⁶⁺, which results in a large electron affinity. This suggests that materials consisting of extraordinarily high-valence cations alongside a wide band gap are promising candidates for the n-type transparent semiconductors. Hence, this study selected TeO₃ as a research target owing to its Te⁶⁺ valence state. For β -type TeO₃, following the potential identified using first-principles calculations [1], we have experimentally demonstrated its optoelectronic properties.

First, we performed electronic structure calculations for β -TeO₃. The result predicted that this material possesses a wide band gap exceeding 3 eV and an ultrahigh ϕ of up to ~ 8 eV. Furthermore, native defect calculations suggest that the formation of electron-trapping oxygen vacancies can be sufficiently suppressed. These results indicate that β -TeO₃ has high potential as an n-type transparent semiconductor for efficient hole injection.

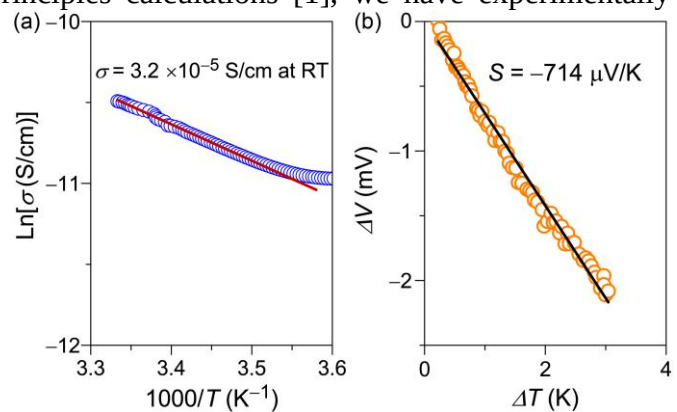


Figure 1: (a) σ - T and (b) thermoelectric motive force (ΔV) plots for β -TeO₃. Solid lines are the fitting results.

Then, a single-phase polycrystalline β -TeO₃ sample was successfully prepared through the thermal decomposition of H₆TeO₆, followed by a purification process. From a diffuse reflectance measurement, the band gap of β -TeO₃ was determined to be 3.1 eV. Figure 1(a) shows the temperature (T) dependence of electrical conductivity (σ). At room temperature (RT), the undoped β -TeO₃ bulk exhibits $\sigma = 3.2 \times 10^{-5}$ S/cm. The σ decreased with decreasing T , yielding a carrier thermal activation energy of ~ 180 meV. Figure 1(b) represents the thermoelectric motive force (ΔV) at RT, where a negative slope with Seebeck coefficient of -714 $\mu\text{V/K}$ proves the n-type nature. These results unambiguously validated that β -TeO₃ is a novel transparent n-type semiconductor. Regarding the carrier generation mechanism, our first-principles calculations suggested that hydrogen at the interstitial sites (H_i), rather than the oxygen vacancy, is a potential shallow n-type donor. Thermal desorption spectroscopy revealed a hydrogen concentration of 0.1 at%, supporting that unintentionally incorporated H_i acts as a donor in the undoped β -TeO₃ polycrystalline sample.

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PLD Growth of ZnO/Mg_xZn_{1-x}O Superlattices for Quantum Cascade Laser

Ryota Takahashi¹, Tatsuya Masuda², Toshihiro Sato³,

Norihiko Sekine⁴, Iwao Hosako⁴, Gen-ichi Hatakoshi², Hideomi Koinuma²

¹ *Nihon University, College of Engineering, Department of Electrical and Electronic Engineering*

² *Smart Combinatorial Technology*

³ *Vacuum Products*

⁴ *National Institute of Information and Communications Technology*

We have developed a precise atomic layer control process using pulsed laser deposition (PLD) to realize quantum cascade laser (QCL) devices using wide-bandgap semiconductors of ZnO and Mg_xZn_{1-x}O. Compared to conventional QCL materials such as GaAs and InP, ZnO has a larger bandgap of 3.3 eV [1] and a high LO phonon energy of 72 meV, making it promising for applications in high temperature QCL devices operating above room temperature.

As shown in Figure 1(a), QCL devices have a structure in which an active layer consisting of a multilayer quantum well structure on the order of several nanometers is sandwiched between cladding layers. To obtain high power, it is necessary to increase the number of repeating periods of the superlattice structure, and precise film thickness control over long periods is essential for fabrication using the PLD method. However, in conventional PLD methods, precise control over long periods is difficult due to contamination of the laser introduction viewport, which has been a major challenge in the fabrication of ZnO-based QCL structures [2].

Here, we have introduced a quartz crystal microbalance (QCM) into a PLD apparatus to attempt precise film thickness control of ZnO and Mg_xZn_{1-x}O, and fabricated a QCL structure on ZnO(000 $\bar{1}$) substrate. The fabricated structure was evaluated using cross-sectional TEM and XRD. Figure 1(b) shows a HAADF-STEM image of a ZnO QCL film grown at 600 °C. The film thickness of each period was revealed to be precisely controlled, as designed in Figure 1 (a). The developed PLD process has opened a new path for the fabrication of QCL devices using wide-bandgap ZnO [3].

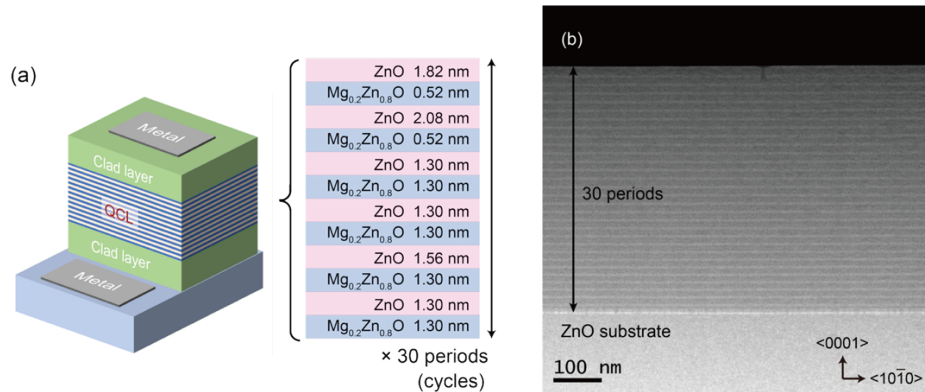


Figure 1: (a) Schematic illustration of the ZnO QCL device structure. (b) HAADF-STEM image of ZnO QCL film.

Acknowledgements

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Calcium Niobate Nanosheet templated monolithic integration of Barium Titanate thin films for Silicon Nitride Integrated Photonics

Ojas Shanker¹, Ruben Dragland², Işık Tarık¹, Hannes Rijckaert¹, Reza Eyni Chenar¹, Minh Duc Nguyen¹, Dennis Meier^{2,3,4}, David Marpaung¹, Gertjan Koster¹, Guus Rijnders¹

¹ MESA+ Institute for Nanotechnology, University of Twente, 7500 AE, Enschede, The Netherlands

² Department of Material Science and Engineering, Norwegian University of Science and Technology, Norway

³ Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Germany

⁴ Research Center Future Energy Materials and Systems, Research Alliance Ruhr, Germany

Barium titanate (BaTiO_3 , BTO) is a promising material platform for next-generation integrated photonics, combining a large electro-optic response with compatibility for high-speed, low-loss photonic architectures. A-axis-oriented BTO thin films are particularly attractive for electro-optic modulation via the in-plane Pockels coefficient (r_{42}). However, monolithic integration of crystalline BTO with silicon nitride photonic platforms remains challenging due to the use of amorphous cladding layers, which are inherently incompatible with conventional epitaxial oxide growth. In this work, we employ a Calcium Niobate (CNO) nanosheet – Strontium Titanate (STO) buffer layer to overcome this substrate induced crystallinity constraint and enable epitaxial a-axis BTO growth on any technologically relevant photonic substrates.

Structural characterization was performed to evaluate the crystalline quality and templated growth behavior of the BTO heterostructures. High-resolution X-ray diffraction confirms textured a-axis BTO growth, while electron microscopy (TEM, STEM, 4D-STEM, and HAADF) reveals the interplay between interfacial structure, strain accommodation, and defect formation within the CNO/STO/BTO heterostructure. Complementary in-plane and out-of-plane PFM measurements establish correlations between microstructure, ferroelectric domain behavior, and the role of grain boundaries in determining device-relevant functionality.

To evaluate device viability, optical simulations, propagation loss and the electro-optic coefficient were extracted for structures on chip. The combined materials platform demonstrates strong potential as a scalable pathway toward monolithically integrated ferroelectric oxide photonics. These results establish templated oxide integration as a compelling route for future electro-optic and multifunctional photonic device architectures.

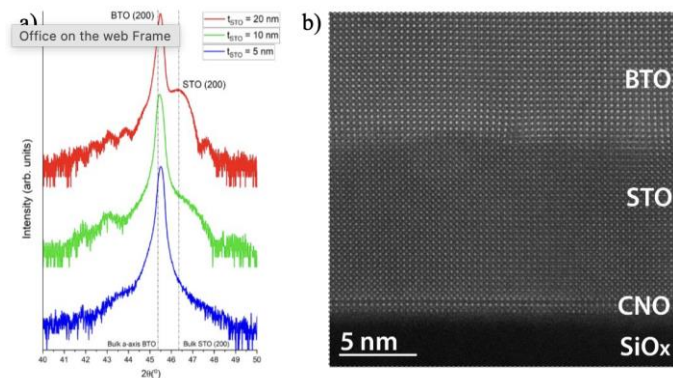


Figure 1: a) a-axis BTO film growth with various STO buffer layer thickness, b) TEM image of the heterostructure.

Epitaxial Synthesis and Physical Properties of BaNbO₃ as a Transparent Conductor

Takanori Shinohara¹, Daichi Oka¹, Shunsuke Miyaji¹, Yasushi Hirose¹

¹ Tokyo Metropolitan University

[Introduction] Transition-metal oxides with strong electronic correlation have recently attracted significant attention as transparent conductors because their plasma frequencies lie in the infrared region [1]. Especially, epitaxial thin films of perovskite-type SrNbO₃ exhibit high optical transmittance extending into the ultraviolet region with a low electrical resistivity of $2.82 \times 10^{-5} \Omega \text{ cm}$ [2,3]. In BaNbO₃, where the *A*-site Sr is substituted by Ba, it is expected that rotational distortions and orbital hybridization from the *A*-site are suppressed, which could alter the magnitude of visible-light absorption derived from the Nb d-d transitions. However, its optical properties have yet to be reported. In this study, we synthesized high-quality BaNbO₃ epitaxial thin films and evaluated their electrical and optical properties.

[Experiment] BaNbO₃ thin films were synthesized by pulsed laser deposition. A Ba₂Nb₂O₇ target was irradiated with a KrF excimer laser pulse (wavelength: 248 nm) inside a high-vacuum chamber with a base pressure of $\sim 4.0 \times 10^{-9}$ Torr. The thin films were deposited on KTaO₃ (001) single-crystal substrates heated at 630–700 °C. The laser energy and repetition rate were set to 7.5 mJ and 4 Hz, respectively. The crystal structure was examined using X-ray diffraction (XRD), while the optical and electrical properties were investigated using a ultraviolet-visible-near-infrared spectrometer and the van der Pauw technique, respectively, at room temperature.

[Results and Discussion] Only the 00*l* peaks were observed in XRD θ –2 θ patterns, while a spot 103 asymmetric diffraction peak was detected in reciprocal space mapping, for the BaNbO₃ thin films. These results indicated successful epitaxial growth of BaNbO₃ despite lattice relaxation from the substrate. Furthermore, the presence of clear Laue fringes around the 002 peak in the high-resolution XRD pattern indicated good crystallinity and a flat surface. The room-temperature electrical resistivity was as low as $5.57 \times 10^{-5} \Omega \text{ cm}$, demonstrating excellent electrical conductivity. The slightly higher resistivity compared to that of SrNbO₃ can be attributed to the larger lattice constant and increased defect density due to the lattice relaxation from the substrate. On the other hand, the absorption coefficient α was smaller than that of SrNbO₃ in the photon energy region below 3 eV (Figure 1). Consequently, a high average internal transmittance of 88% was achieved in the visible-light region (wavelength: 400–700 nm) for a 20-nm-thick film (sheet resistance 28: Ω/sq). In the presentation, we will also discuss the film thickness dependence of the electrical and optical properties.

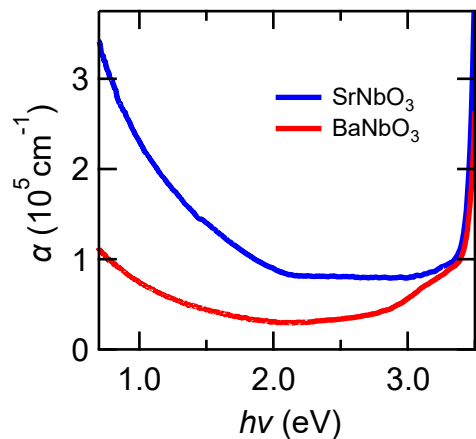


Figure 1. Absorption coefficient spectrum of the BaNbO₃ epitaxial thin film. The spectrum reported for SrNbO₃ epitaxial thin film is also plotted for comparison [3].

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Ce-Doped BaZrO₃ as a Large-Lattice-Parameter Substrate Platform

Yoon Seok Oh¹

¹*Department of Physics, UNIST, Ulsan 44919, Republic of Korea*

Among widely used cubic perovskite oxide substrates, BaZrO₃ has an exceptionally large lattice parameter of approximately 4.19 Å. This large lattice parameter enables substantial tensile strain to be imposed on overlying perovskite films, allowing the epitaxial stabilization of functional oxide phases that are difficult to access on conventional perovskite substrates [1]. Nevertheless, for several intriguing perovskite oxides, including superconducting Ba(Bi,Sb)O₃ with a lattice parameter of approximately 4.3–4.5 Å, even BaZrO₃ remains insufficient to provide an ideal lattice match, and the residual mismatch can still hinder high-quality epitaxial growth. To overcome this limitation, we have explored Ce substitution as a route to expand the BaZrO₃ lattice beyond 4.19 Å. In the BaZr_{1-x}Ce_xO₃ system, incorporation of Ce on the Zr site is expected to increase the average B-site ionic radius and thereby enlarge the perovskite lattice, while also driving structural instability and possible phase evolution at higher Ce contents. In this poster, we present our experimental efforts to synthesize Ce-doped BaZrO₃ and examine how Ce incorporation modifies the lattice parameter, phase stability, and structural symmetry of the BaZrO₃-based perovskite system. These results provide a materials-design pathway toward large-lattice-parameter oxide substrates for tensile-strain engineering of emerging functional and superconducting perovskite films.

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Observation of Kink Dispersion in Antiferromagnetic Kondo-lattice CePtAl₄Ge₂

Jeonghwan Oh¹, Younsik Kim², Jinyoung Kim², Sehoon Kim², Soohyeon Shin^{3,4}, Elio Vescovo⁵, Sangjae Lee⁶, Charles H. Ahn⁶, Changyoung Kim², Tuson Park⁴, Bogyu Jang⁷, and Byungmin Sohn¹

¹ Department of Physics, Sungkyunkwan University, Suwon 16419, Korea

² Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea

³ PSI Center for Neutron and Muon Sciences, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

⁴ Center for Quantum Materials and Superconductivity (CQMS) and Department of Physics, Sungkyunkwan University, Suwon 16419, Korea

⁵ National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, New York 11973, USA

⁶ Department of Applied Physics, Yale University, New Haven, CT 06520, USA

⁷ Department of Materials Science and Engineering, Kyung Hee University, Yongin 17104, Korea

Heavy-fermion behavior has been widely reported in various strongly correlated systems, including transition-metal oxides, but the canonical realization remains the Kondo lattice in 4f compounds^[1]. CePtAl₄Ge₂ (CPAG) is an antiferromagnetic Kondo-lattice system that has attracted much interest due to the Kondo effect, which originates from the hybridization between itinerant Pt conduction electrons and localized Ce 4f electrons. While the magnetic and electronic transport properties of single-crystal CPAG have been extensively characterized^[2, 3], its electronic band structure has not yet been reported. In this work, we investigate the low-energy band dispersion of CPAG using high-resolution (1 meV) laser-based angle-resolved photoemission spectroscopy (ARPES) at a base temperature of $T = 5.8$ K, within the reported Kondo coherence regime ($T^ = 6$ K). We observe clear renormalization of the conduction band near the Fermi level with a characteristic energy scale of about 5 meV, consistent with Kondo hybridization between the Ce 4f moments and conduction electrons. These results provide the first direct observation of Kondo-driven band renormalization in CPAG. Notably, given the exceptionally low coherence temperature ($T^* = 6$ K) reported for this compound, resolving such subtle renormalization features required the superior energy resolution of laser-ARPES, establishing CPAG as one of the rare Kondo-lattice systems in which coherent Kondo hybridization has been directly visualized deep in the extreme low- T^* regime and paving the way for momentum-resolved studies of the electronic structure in weakly hybridized heavy-fermion compounds.*

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32nd International Workshop on Oxide Electronics

iWOE32 - Sendai, Japan

Poster Session B

(Sessions 4, 5, 6)

Date & Time : September 29, 2026 (Tue) 14:00 - 16:00

Venue : Main Hall / Poster Area

Intrinsic low-spin state and strain-tunable anomalous Hall scaling in high-quality SrRuO₃ (111) films

○Harunori Shiratani,^{1,2} Yuki K. Wakabayashi,¹ Yoshiharu Krockenberger,¹ Masaki Kobayashi,¹ Kohei Yamagami,³ Takahito Takeda,⁴ Shinobu Ohya,^{2,5} Masaaki Tanaka,^{2,5} and Yoshitaka Taniyasu¹

¹ Basic Research Laboratories, NTT, Inc., ² Department of Electrical Engineering and Information Systems, The University of Tokyo, ³ Japan Synchrotron Radiation Research Institute (JASRI), ⁴ Department of Chemical System Engineering, The University of Tokyo, ⁵ Center for Spintronics Research Network, The University of Tokyo

SrRuO₃ (SRO) is a 4d transition-metal oxide perovskite that has been extensively studied as a prototypical itinerant ferromagnet with intermediate electron correlation and large magnetocrystalline anisotropy [1]. Also, recent observation of quantum transport phenomena of Weyl fermions in SRO has attracted considerable interest as a magnetic Weyl semimetal [2]. Moreover, (111)-oriented SRO has buckled honeycomb-like surface/interface structures. There have been reports of a strain-induced high-spin state ($4 \mu_B$ per Ru⁴⁺) on the (111) plane [3], prompting a long-standing debate over its existence. Motivated by such prospects, considerable effort has been devoted to improving SRO (111) film quality [4]. Nevertheless, the SRO (111) surface is thermodynamically unstable due to its high polarity and large bond density per unit area, which has long hampered the epitaxy of high-quality SRO (111) films. Therefore, there is a strong need to fabricate high-quality SRO (111) films and to systematically elucidate the correlations between their structure, magnetism, and transport properties.

In this study, we grew high-quality SRO (111) films with thicknesses $t = 1.2\text{--}60$ nm on SrTiO₃ (STO) (111) by machine-learning-assisted molecular beam epitaxy and characterized their structure, magnetism, and magnetotransport property [5]. X-ray diffraction reciprocal space mapping (HRXRD-RSM) and cross-sectional scanning transmission microscopy (STEM) confirmed epitaxy with in-plane coherent compressive strain for $t = 10$ and 20 nm, and strain relaxation at $t = 60$ nm. The coherently strained film with $t = 20$ nm was grown epitaxially with an abrupt substrate/film interface [Fig. 1(a)]. The 60-nm-thick film reached a residual resistivity ratio (RRR) of 45.5, the highest value reported for SRO (111) films. We observed robust, non-saturating linear positive magnetoresistance (MR) up to 14 T and analyzed the anomalous Hall effect (AHE) by scaling to separate intrinsic (Karplus–Luttinger) and extrinsic (side-jump) contributions. Comparison between coherently strained ($t = 10, 20$ nm) and relaxed ($t = 60$ nm) films shows that (111) epitaxial strain systematically tunes the relative weight of the AHE mechanisms. Superconducting quantum interference device (SQUID) magnetometry and Ru $M_{2,3}$ and O K -edge soft X-ray absorption spectroscopy (XAS)/X-ray magnetic circular dichroism (XMCD) together with sum-rule analysis indicated an intrinsically low-spin Ru ground state for both coherently strained and relaxed films; Both coherently strained and relaxed films have a magnetic moment below $1.5 \mu_B/\text{Ru}$ [Fig. 1(b)]. These results position SRO (111) as a materials platform for (111)-oriented oxide heterostructures and spin-topological devices.

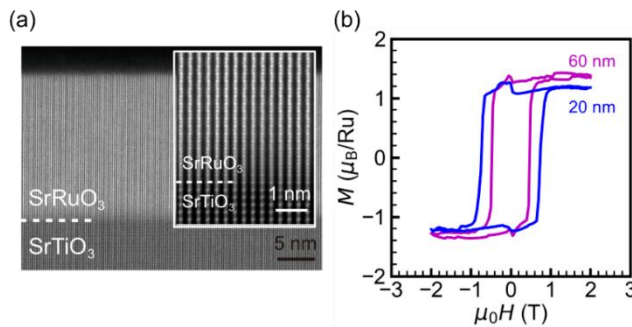


Figure 1 (a) STEM image of the SRO (111) film with $t = 20$ nm viewed along the $[11\bar{2}]$ direction. The inset shows the magnified, high-resolution image near the interface. (b) M - $\mu_0 H$ curves for the SRO (111) with $t = 20$ nm at 2.5 K and $t = 60$ nm at 4 K. A magnetic field was applied perpendicular to the film plane.

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Higer-order contributions in anomalous Hall effect as revealed by full-angular field-direction measurements of SrRuO₃ ultrathin films

Haruto Kaminakamura¹, Naohiro Tezuka¹, Shinichi Nishihaya¹, Tetsuya Furukawa², Motoi Kimata³, Mitsuru Akaki⁴, Yuki Deguchi¹, Masaki Uchida^{1,5}

¹ Dept. of Physics, Institute of Science Tokyo

² Institute for Materials Research (IMR), Tohoku University

³ Japan Atomic Energy Agency (JAEA)

⁴ Core Facility Center, Institute for Academic Research and Co-Creative Innovation (ARC), Kobe University

⁵ Toyota Physical and Chemical Research Institute (TPCRI)

In magnetic materials, anomalous Hall effect (AHE) under out-of-plane magnetic field has been primarily studied. Recently, in-plane AHE induced by in-plane magnetic field has been reported and has attracted attention as a new symmetry-constrained transport phenomenon that reflects the quantum geometry of the band structure [1]. In the previous work reporting spontaneous in-plane AHE in (111) SrRuO₃ ultrathin films [2], we have revealed that an out-of-plane orbital ferromagnetic state is realized, in which finite out-of-plane orbital magnetization remains even when spin magnetization lies within the plane. However, the origin of the nonmonotonic polar angle θ dependence of Hall resistivity observed under increasing magnetic fields remains unresolved.

In this study, we have performed full-angular field-direction measurements under higher magnetic fields using a 25 T cryogen-free superconducting magnet (25T-CSM) at Institute for Materials Research (IMR), measuring the polar-angle θ dependence of the Hall resistivity while rotating the azimuthal angle φ in 5° steps. These measurements reveal that the magnitude of in-plane AHE becomes comparable to that of out-of-plane AHE at 24 T. Fitting analyses assuming (111)-oriented trigonal and cubic systems further demonstrate that the nonmonotonic polar angle θ dependence originates from the higher-order terms allowed by trigonal symmetry.

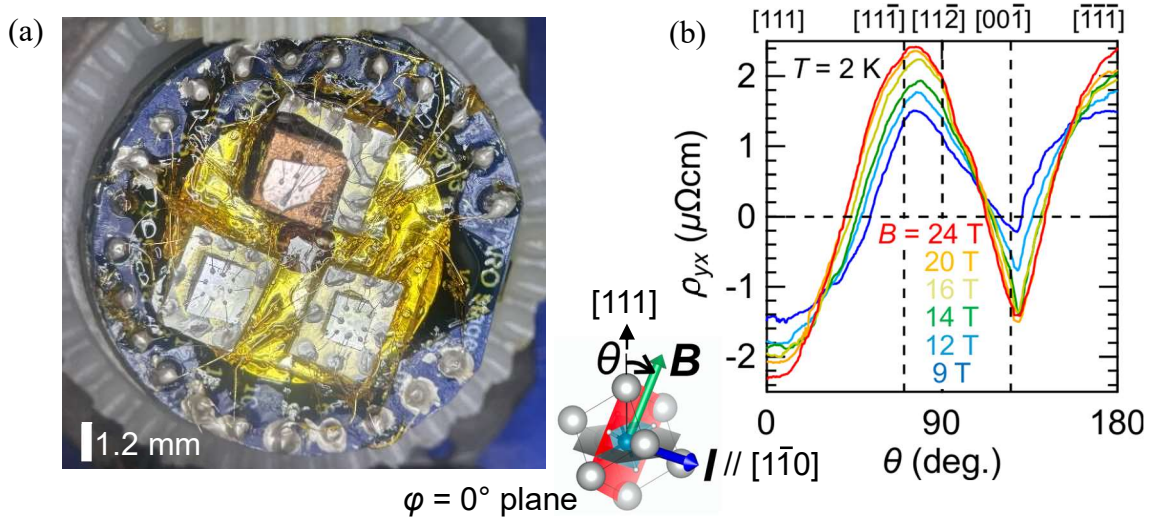


Figure 1: (a) Experimental setup for full-angular field-direction measurements of (111) SrRuO₃ ultrathin films on the two-axis rotating probe. (b) Polar angle dependence of Hall resistivity in the $\varphi = 0^\circ$ plane at high magnetic fields.

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Anisotropic Magnetoresistance and Magnetic Field-Tunable Weyl Nodes in Weyl metal SrRuO₃ Thin Films

Akhilesh Kr. Singh¹, Uddipta Kar¹, Elisha Cho-Hao Lu¹, P. V. Sreenivasa Reddy², Fu-En Cheng¹, Wazid Ahmed¹, Song Yang³, Chun-Yen Lin³, Chia-Hung Hsu³, Guang-Yu Guo^{2,4} and Wei-Li Lee¹

¹ *Institute of Physics, Academia Sinica, Taipei, Taiwan*

² *Department of Physics, National Taiwan University, Taipei, Taiwan*

³ *National Synchrotron Radiation Research Center, Hsinchu, Taiwan*

⁴ *Physics Division, National Center for Theoretical Sciences, Taipei 10617, Taiwan*

Magnetic Weyl semimetals offer an ideal playground for exploring the interplay between topology, magnetism, and electronic correlations. Realizing these emergent phenomena in thin films requires high crystal quality and control over structural domains [1-4]. In this work, untwinned monoclinic SrRuO₃ (SRO) thin films were grown on SrTiO₃ (001) substrates using the adsorption-controlled growth technique [4,5]. The as-grown film with a thickness of about 13.7 nm yields an exceptionally high residual resistivity ratio (RRR $\sim$ 24) and low residual resistivity of about 9 $\mu\Omega\text{cm}$. By patterning the SRO thin film into a sunbeam-shaped device layout, rigorous angle-dependent magnetotransport measurements with various in-plane field-orientation can be performed to investigate the chiral anomaly effect [6]. At low temperatures ($T \leq 25$ K), we uncover unusual angular dependencies in the in-plane magnetoresistance and Hall signals, marked by a prominent fourfold-symmetric component. These transport characteristics match electronic band structure calculations, which reveal that manipulating the magnetization orientation dramatically shifts the energetic positions of the Weyl nodes relative to the Fermi level. Rotating the magnetization away from the principal crystalline axes moves the closest Weyl nodes farther from the Fermi surface, suppressing the chiral anomaly response. This study successfully demonstrates that field-driven magnetization rotation can effectively tune robust topological transport states in high-quality oxide thin films.

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Thickness-Dependent Suppression of Near-Fermi-Level Density of States in Ultrahigh-Quality SrRuO₃ Thin Films

Masaki Kobayashi^{1,2,3,*}, Yuichi Seki³, Takahito Takeda⁴, Shin-ichi Fujimori⁵, Atsushi Fujimori⁶, Yoshiharu Krockenberger¹, Yuki K. Wakabayashi^{1,†} and Yoji Kunihashi¹

¹Basic Research Laboratories, NTT, Inc., Atsugi, Kanagawa, Japan

²Center for Spintronic Research Network, University of Tokyo, Hongo, Tokyo, Japan

³Department of Electrica Engineering and Information Systems, University of Tokyo, Hongo, Tokyo, Japan

⁴Department of Chemical System Engineering, University of Tokyo, Hongo, Tokyo, Japan

⁵Materials Science Research Center, Japan Atomic Energy Agency, Sayo-gun, Hyogo, Japan

⁶Department of Physics, University of Tokyo, Hongo, Tokyo, Japan

Email: *masaki.kobayashi.brl@ntt.com, †yuuki.wakabayashi@ntt.com

Transition-metal ferromagnetic oxide SrRuO₃ (SRO) has been enthusiastically studied for more than 60 years because of its singular physical properties [1,2]. In particular, thanks to the state-of-the-art crystal growth technique [3], quantum transport phenomena of Weyl fermions have recently been observed in ultrahigh-quality SRO films [4,5]. Reducing the thickness is expected to enhance surface and interface scattering and to drive the system toward a metal-insulator transition (MIT), while recent X-ray magnetic circular dichroism studies showed robust ferromagnetism even in ultrathin SRO films [6,7]. Here, to understand the microscopic origin of the thickness-driven suppression of metallicity in SRO, we use soft X-ray photoemission spectroscopy to clarify how the density of states (DOS) near Fermi level (E_F) evolves with thickness in ultrahigh-quality SRO thin films.

In transition-metal oxides, which are strongly correlated electron systems, MIT often involves an interplay between disorder and electron-electron correlation. For example, Altshuler and Aronov predicted that weak electron-electron interaction in disordered metallic medium leads to a singularity of DOS $[N(E)]$ near E_F proportional to $|E - E_F|^{1/2}$ [8,9]. More generally, $N(E)$ reflected electronic interaction toward MIT can be expressed as $N(E) \propto |E - E_F|^x$, where x depends on type of electronic interaction, dimensionality, and conductivity of the systems.

SRO films grown by molecular beam epitaxy with thicknesses of 1, 2, 3, 4, 5, and 60 nm were measured at BL23SU, SPring-8. The valence-band (VB) spectra in Fig. 1 show a monotonic depletion of spectral weight around E_F with decreasing thickness from 4 nm to 1 nm. The coherent peak near E_F looks disappears in the films with thickness less than 3 nm, indicating the conduction electrons lose their coherence in the ultrathin region. The exponent x for depletion of $N(E)$ was estimated as $x \sim 1$ for the 1 nm film. Its nearly linear power-law behavior, together with the absence of an energy gap, suggests disorder-induced DOS suppression due to enhancement of surface and interface scattering in the ultrathin limit. These results provide a microscopic picture of how SRO approaches a thickness-controlled MIT while retaining metallic character down to 1 nm.

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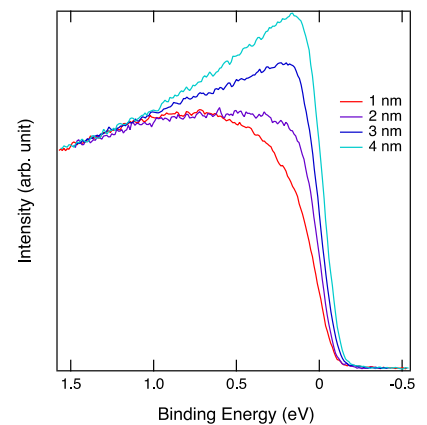


Fig. 1. Thickness-depenent VB spectra of ultrathin SRO films (1-4 nm).

Quantitative determination of the Dzyaloshinsky-Moriya interaction in SrIrO₃/SrRuO₃ bilayers by tilted-field full-droplet analysis

Karl Frischmuth¹, Junichi Shiogai^{1,2}, Kohei Ueda^{1,2,3}, Takamasa Usami^{2,3}, Kohei Hamaya^{2,3,4}, Takanori Kida⁵, Masayuki Hagiwara⁵, and Jobu Matsuno^{1,2,3}

¹*Department of Physics, Graduate School of Science, The University of Osaka, Japan*

²*Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives, The University of Osaka, Japan*

³*Center for Spintronics Research Network, Graduate School of Engineering Science, The University of Osaka, Japan*

⁴*Department of Systems Innovation, Graduate School of Engineering Science, The University of Osaka, Japan*

⁵*Center for Advanced High Magnetic Field Science, Graduate School of Science, The University of Osaka, Japan*

The Dzyaloshinskii–Moriya interaction (DMI) is a key mechanism for stabilizing chiral spin textures such as skyrmions. Although DMI has been extensively studied in different material systems, its quantitative evaluation in oxide heterostructures remains limited, as most previous studies have relied on indirect probes such as the topological Hall effect (THE) and magnetic force microscopy. SrIrO₃/SrRuO₃ bilayers are a notable example, exhibiting hump-like Hall anomalies that have often been discussed in connection with THE [1,2]. This motivates an independent and quantitative evaluation of DMI in this system.

Here, we measure the coercivity of epitaxial SrIrO₃/SrRuO₃ (SIO/SRO) bilayers grown on SrTiO₃ (STO) under tilted magnetic fields. We investigate SIO(2 u.c.)/SRO bilayers with various SRO thicknesses together with a reference SRO (6 u.c.)/STO film. The results are analyzed using the magnetic full-droplet model to quantitatively evaluate DMI [3]. By fitting the critical in-plane field component H_{DMI} , we extract the DMI coupling strength D , enabling quantitative evaluation independent of skyrmion-stability assumptions. The extracted H_{DMI} values are significantly larger in all bilayer samples than in the single-layer film, revealing a pronounced interfacial DMI originating from the SIO/SRO interface. The observed thickness dependence of the extracted DMI values is broadly compatible with the interfacial DMI picture. These results provide a quantitative basis for assessing interfacial DMI in oxide heterostructures, highlighting the importance of the SIO/SRO interface in chiral spin textures.

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Enhanced Electric-Field Control of Spin Waves through Lu-Induced Lattice Distortion in Garnet Oxides

Univ. Tokyo, ° Kamrul Hasan*, Md Shamim Sarker, E M K Ikbali Ahamed, Ritsuki Sonetaka, Hiroyasu

Yamahara, Munetoshi Seki, and Hitoshi Tabata

*E-mail : kamrulhira@g.ecc.u-tokyo.ac.jp

Spin waves (SWs) are promising information carriers for beyond-CMOS computing owing to their wave-based signal processing capability and ultra-low energy dissipation. Electric-field manipulation of SW propagation has attracted considerable attention as an energy-efficient alternative to current-driven control methods; however, the achievable tunability remains limited in conventional yttrium iron garnet (YIG) systems due to weak magnetoelectric coupling [1,2]. Here, we demonstrate enhanced electric-field-induced SW frequency modulation in Lu-substituted yttrium iron garnet, $Y_{3-x}Lu_xFe_5O_{12}$ (YLuIG), grown on SGGG substrates. By systematically varying the Lu concentration, the lattice distortion of the garnet film is increased, resulting in significantly improved electric-field responsiveness. A maximum frequency shift of approximately 9 MHz is achieved at 400 kV/cm for $Y_{0.75}Lu_{2.25}$ IG, nearly twice that of undoped YIG, demonstrating the effectiveness of compositional strain engineering for low power magnonic applications.

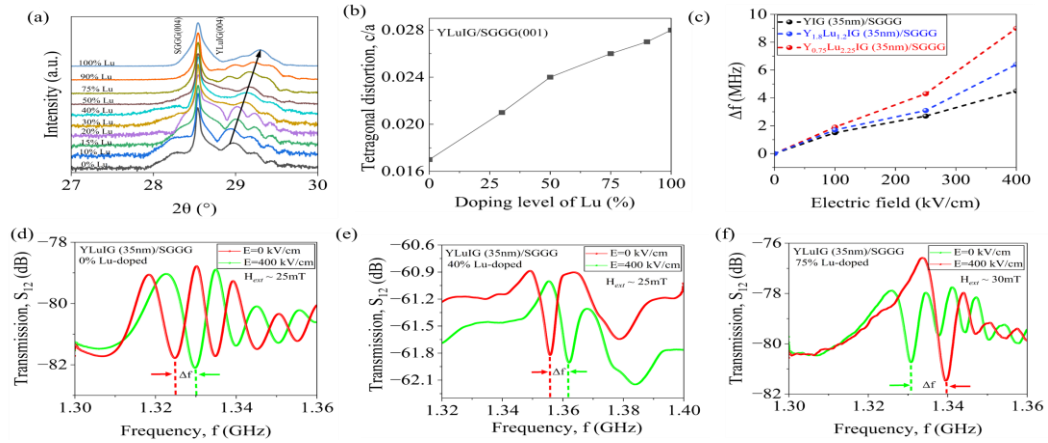


Figure 1. (a) X-ray diffraction patterns of YLuIG thin films with varying Lu concentrations. (b) Dependence of the tetragonal distortion (c/a) on Lu doping level. (c) Frequency shift (Δf) as a function of electric field for YIG and Lu-doped YLuIG thin films. Transmission spectra measured at $E = 0$ and 400 kV/cm for (d) undoped YIG, (e) 40% Lu-doped YLuIG, and (f) 75% Lu-doped YLuIG films, showing electric-field-induced resonance-frequency shifts.

A 35-nm-thick PLD-grown YLuIG film was used as the spin-wave medium, with three coplanar waveguide (CPW) antennas for SW excitation and detection. An interdigital electrode (IDE) was integrated along the $+k$ propagation path to enable electric-field control of SW propagation. X-ray diffraction measurements reveal a systematic shift of the YLuIG(004) peak with increasing Lu concentration, as shown in Fig. 1(a). The corresponding tetragonal distortion increases monotonically with Lu content [Fig. 1(b)], indicating enhanced lattice deformation in the films. The increased distortion is expected to promote inversion-symmetry breaking and strain-induced orbital polarization, thereby strengthening the coupling between electric fields and magnetic anisotropy. Consequently, the electric-field sensitivity of the spin-wave dispersion is enhanced in Lu-substituted films. The electric-field-induced spin-wave frequency modulation was evaluated from transmission spectra measured at $E = 0$ and 400 kV/cm for the 0%, 40%, and 75% Lu-doped samples [Fig. 1(d–f)]. The frequency shift (Δf), extracted from the displacement of the resonance minima, increases with both electric-field strength and Lu concentration, as summarized in Fig. 1(c). In particular, the 75% Lu-doped film exhibits a frequency shift of approximately 9 MHz at 400 kV/cm, nearly twice that of undoped YIG. These results demonstrate that Lu-induced tetragonal distortion effectively enhances magnetoelectric coupling and enables stronger electric-field control of spin waves.

Acknowledgement

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Sub-nanosecond spin dynamics in yttrium iron garnet-based spin-frustrated thin films

Ritsuki Sonetaka¹, Dongxun Yang¹, Ruyi Dai¹, MD Shamim Sarker¹, Hiroyasu Yamahara¹, Hitoshi Tabata¹

¹University of Tokyo, Japan

Yttrium iron garnet (YIG) is a prototypical insulating ferrimagnet for studying low-loss magnetization dynamics due to its low magnetic damping[1]. While such coherent dynamics is attractive for spin-wave-based devices, nonlinear dynamical responses are also important for nonconventional computing applications[2]. A possible route to realize such responses is chemical substitution, which can modify local structural and magnetic environments. In particular, Co and Si substitution can enhance magnetic anisotropy and introduce local disorder through the modified Fe-site environment[3]. In this study, we investigate the dynamic magnetic properties of YIG ($\text{Y}_3\text{Fe}_5\text{O}_{12}$) and Co- and Si-substituted YIG ($\text{Y}_3\text{Fe}_{4.6}\text{Co}_{0.2}\text{Si}_{0.2}\text{O}_{12}$), grown on GGG ($\text{Gd}_3\text{Ga}_5\text{O}_{12}$) (100) substrates. Microwave reflection measurements shown in Fig. 1(a) revealed two V-shaped magnetic resonance branches in the reflection S_{11} spectrum of $\text{Y}_3\text{Fe}_{4.6}\text{Co}_{0.2}\text{Si}_{0.2}\text{O}_{12}$. To confirm these microwave resonance features in the time domain, time-resolved magneto-optical Faraday effect (TR-MOFE) measurements were performed on the same film (Fig. 1(b)). The observed oscillatory signal was well reproduced by a superposition of two damped oscillations $A_1 e^{-t/\tau_1} \sin(2\pi f_1 t) + A_2 e^{-t/\tau_2} \sin(2\pi f_2 t)$, whose frequencies correspond to the two resonance branches observed in the S_{11} spectrum. To discuss the origin of this two-frequency resonance, angle-dependent ferromagnetic resonance (FMR) was performed. Co and Si substitution increased the effective magnetization $4\pi M_{\text{eff}}$ from 2092 Oe for YIG to 2731 Oe for $\text{Y}_3\text{Fe}_{4.6}\text{Co}_{0.2}\text{Si}_{0.2}\text{O}_{12}$, accompanied by an increase in the magnitude of the effective perpendicular anisotropy K_u from -7.8×10^3 erg/cm³ to -4.6×10^4 erg/cm³. These results suggest that the enhanced magnetic anisotropy and local disorder due to Co and Si substitution may lead to the formation of magnetic domains or inhomogeneous magnetic regions with different resonance conditions. Such multi-mode magnetization dynamics may be useful for future non-conventional computing applications, such as reservoir computing, where complex nonlinear dynamics is favored.

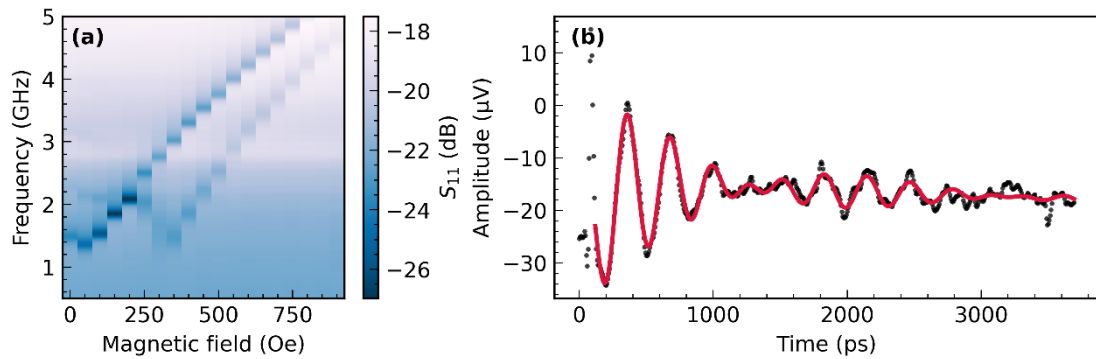


Figure 1: Magnetization dynamics of $\text{Y}_3\text{Fe}_{4.6}\text{Co}_{0.2}\text{Si}_{0.2}\text{O}_{12}$ thin film. (a) Magnetic-field dependence of the S_{11} spectrum. (b) TR-MOFE signal fitted by a superposition of two damped oscillations.

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Ultrafast Spin Dynamics in Strain-induced Spin Frustrated Lutetium Iron Garnet (LuIG) Films on YAG Revealed by Time Resolved Magneto-Optic Faraday Effect

Dongxun Yang¹, Md Shamim Sarker², Hiroyasu Yamahara¹, Munetoshi Seki^{1,2}, Hitoshi Tabata^{1,2}

¹ Department of Bioengineering, Graduate School of Engineering, The University of Tokyo

² Department of Electrical Engineering and Information Systems, Graduate School of Engineering, The University of Tokyo

Magnetic anisotropy in epitaxial garnet films is highly sensitive to strain, structural relaxation, and film thickness, providing a route to control ultrafast spin dynamics[1]. In this research, we investigate thickness-dependent spin precession in Lutetium Iron Garnet (LuIG) thin films on YAG using time-resolved magneto-optical Faraday effect (TR-MOFE). Representative time-domain signals in Fig. 1(a) and the corresponding FFT spectra in Fig. 1(b) reveal coherent magnetization precession whose frequency and spectral character evolve strongly with film thickness. XRD and reciprocal space mapping show thickness-dependent structural evolution and strain relaxation (Figs. 1(c) and 1(f)). Magnetic-field-angle-dependent measurements in Fig. 1(d) further show distinct frequency evolution among different thicknesses, which is analyzed using the Smit-Belgers formalism[2], based on magnetic free energy. While thinner films are well described by a conventional effective magnetization term, thicker films require higher-order anisotropy contributions to reproduce the observed frequency evolution. The extracted anisotropy parameters in Fig. 1(e) show that the second-order perpendicular anisotropy (K_2) increases markedly with thickness, accompanied by a sign change of the effective magnetization in the thickest film. These results indicate that strain relaxation generates an inhomogeneous magnetoelastic anisotropy landscape, producing competing local easy axes and spin frustration.

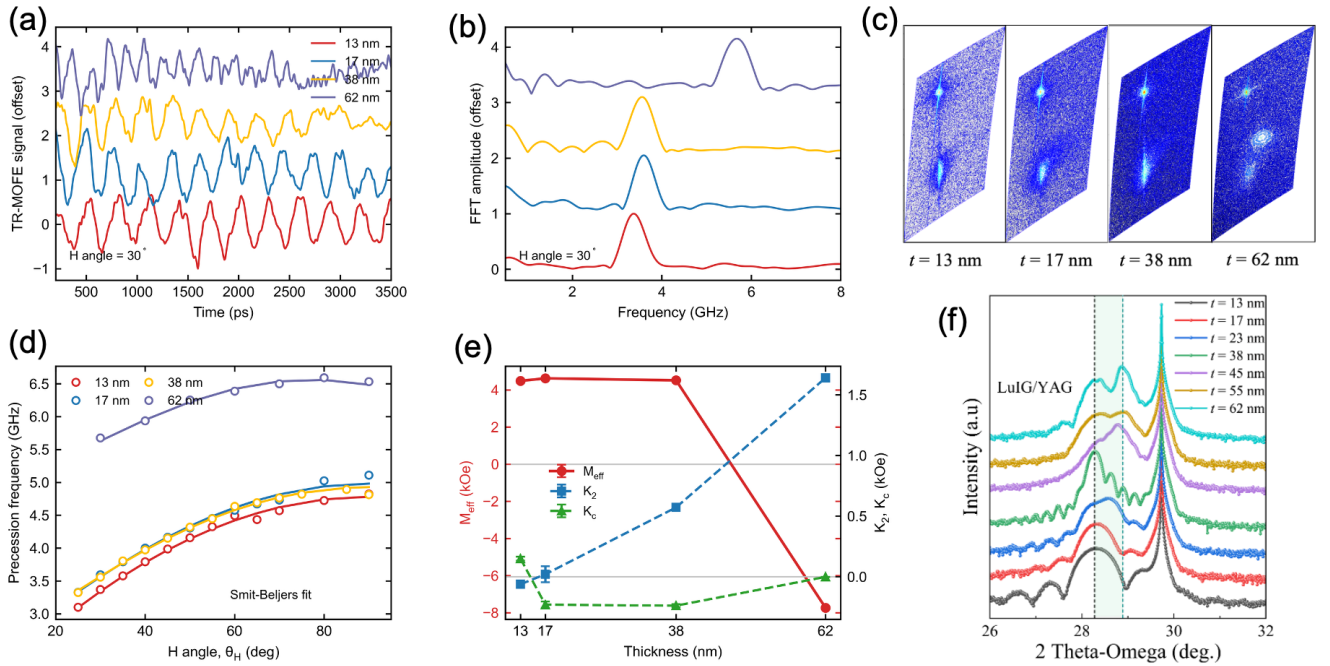


Figure 1. Thickness-dependent spin dynamics and magnetic anisotropy in LuIG/YAG films. (a) Representative TR-MOFE signals measured at $\theta_H = 30^\circ$. (b) Corresponding FFT spectra. (c) Reciprocal space maps for selected film thicknesses. (d) Magnetic-field-angle dependence of the precession frequency fitted by the Smit-Belgers model. (e) Extracted M_{eff} , K_2 , and K_c as a function of film thickness. (f) XRD $2\theta - \omega$ scans showing thickness-dependent structural evolution.

This research was supported by Institute for AI and Beyond for the University of Tokyo, JST, CREST Grant Number JPMJCR22O2, Japan, AMED under Grant Number JP22zf0127006, JSPS KAKENHI Grant Number 25H00730, 25H02602, 25K22084, Tateisi Science and Technology Foundation.

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Nonlinear optical imaging of magnon transport in oxide magnetic insulators

G. De Luca¹

¹ Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Carrer dels Til·lers, 08193 Cerdanyola del Vallès, Spain

Complex oxide magnetic insulators offer an extraordinary playground for spin transport physics, combining rich magnetic ground states, tunable topology, and epitaxial compatibility with functional heterostructures [1]. Among these, ferromagnetic pyrochlore oxides stand out as a largely unexplored family with strong potential for magnon spintronics [2]. Yet the tools needed to observe and spatially map spin transport in these materials remain underdeveloped. Conventional detection schemes, based on platinum/oxide heterostructures and inverse spin Hall voltage measurements, provide spatially averaged signals that obscure the local physics one would like to resolve.

Nonlinear optics offers a different path. Magnetization-induced Second Harmonic Generation (MSHG) is intrinsically sensitive to broken time-reversal symmetry at magnetic interfaces, couples directly to the local order parameter, and delivers sub-micron spatial resolution [3,4]. These properties make it an ideal probe for imaging the non-equilibrium magnon populations generated by magnon transport processes such as the Spin Seebeck, Spin Nernst, and Magnon Hall Effects. Crucially, MSHG's interface selectivity is especially powerful in oxide heterostructures, where symmetry breaking and emergent phenomena are concentrated at epitaxial interfaces.

The framework outlined here is to be developed within a recently awarded ERC Starting Grant (TOPOLOGIQ), which aims at using pyrochlore oxide thin films and artificial heterostructure as a designable platform to test these ideas. The goal is to demonstrate spatially resolved maps of magnon transport (without electrical readout) thereby opening a new optical window onto spin caloritronics and topological magnon physics in oxides.

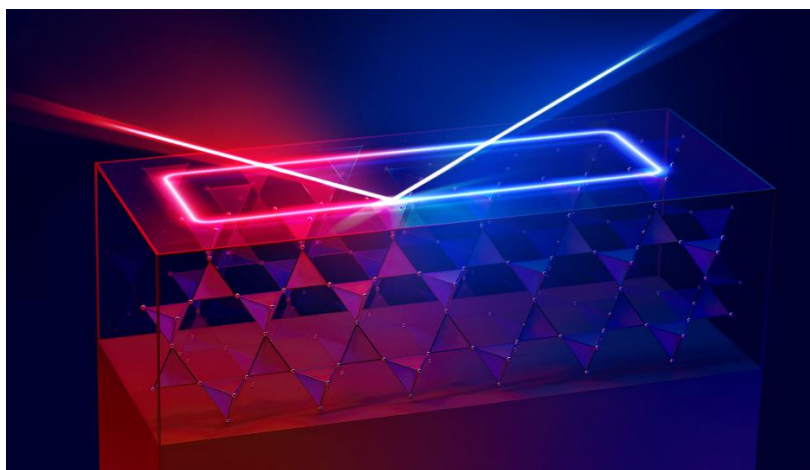


Figure 1: Artistic sketch of a pyrochlore lattice embedded in a thin-film heterostructure. The colored laser beams illustrate the nonlinear optical probe concept

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Orbital Hybridization Induces Giant Cubic Rashba Effect at Cu/WO₃ Interface

Md Aktar Hossain¹, Saikat Das¹

¹Department of Physics, *Indian Institute of Technology Kharagpur, India*

The Rashba spin–orbit interaction (SOI) enables a direct interconversion between charge and spin currents, a key mechanism underpinning spintronic devices [1]. The phenomenon arises from spin-split electronic bands induced by broken inversion symmetry, where the magnitude of the spin splitting directly governs the efficiency of spin-charge interconversion. The ability to electrically tune Rashba splitting thus offers a pathway toward low-power, voltage-controlled spintronic devices. In addition to the conventional linear Rashba effect, recent studies highlight the importance of nonlinear (cubic) Rashba contributions, which play a central role in nonlinear spin–charge conversion and quantum metric effects [2].

Although Rashba SOI has been widely explored at heavy-metal interfaces, light-metal interfaces remain less studied due to their weaker atomic SOI. Surprisingly, recent experiments have revealed notable Rashba effects at Cu/Bi₂O₃ and Py/Bi₂WO₆ interfaces, suggesting that orbital hybridization with heavy elements can enhance spin–orbit coupling [3,4]. Motivated by these observations, we perform first-principles DFT calculations to explore the Rashba effect at the interface between a light metal (Cu) and a heavy-element oxide (WO₃).

Using Cu(001)/WO₃(001) as a model light-metal/heavy-oxide heterostructure, we demonstrate the emergence of a robust interfacial Rashba spin-split state driven by strong W spin–orbit coupling and W–Cu orbital hybridization. First-principles calculations reveal the coexistence of linear and cubic Rashba effects. The cubic Rashba contribution increases with Cu thickness due to electronic cross-coupling between the vacuum/Cu and Cu/WO₃ interfaces, and saturates at larger thicknesses, enabling extraction of the intrinsic Rashba parameters. We obtain a linear Rashba coefficient of $\alpha_R \approx +0.5 \text{ eV}\cdot\text{\AA}$ and a giant cubic Rashba coefficient of $\tilde{\alpha}_R \approx -1.9 \text{ eV}\cdot\text{\AA}^3$. Furthermore, sizable linear and cubic Rashba spin splittings persist across other orientations also Cu(110) and Cu(111), demonstrating the robustness and orientation-independent nature of the effect. These findings establish the Cu/WO₃ interface as a promising platform for Rashba spintronics and nonlinear spin–orbit phenomena, with potential applications in tunable spin–charge interconversion and oxide-based spin–orbitronic devices [5].

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Thin Film Growth of Altermagnetic Candidate α -Fe₂O₃ by Pulsed Laser Deposition

Jack Dorsey¹, Aliya Kaplan¹, Jennifer Fowlie¹

¹ *Northwestern University Department of Materials Science and Engineering*

Hematite (α -Fe₂O₃) is a compelling candidate for both altermagnetic spintronics and magnonics. Altermagnets represent a class of magnetic materials that have fully compensated moments yet have broken time-reversal symmetry and lifted Kramers degeneracy in momentum space leading to spin-split bands, making them attractive for spintronic and magnonic applications [1]. Hematite is distinguished by its complex topological spin-textures, low Gilbert damping, and switchable domains [2-4]. Additionally, it exhibits large spin Hall magnetoresistance alongside long-range, polarized magnon propagation [3-8]. Yet, integrating this material into functional devices requires precise control over its magnetic phase and structural order. Using Pulsed Laser Deposition (PLD), we systematically optimize the growth of α -Fe₂O₃ thin films, focusing particularly on the role of strain in controlling film properties. It was found that substrate temperature, fluence, oxygen pressure and deposition rate play a key role in film strain and quality. Strain controls the magnetic anisotropy energy, tuning the competition between long-range dipole and single-ion anisotropy [9]. By rigorously tuning the oxidation, strain, and film thickness, we achieve high-crystalline-quality films. These high-quality films serve as an ideal platform for probing and tuning α -Fe₂O₃'s fundamental magnetic order and phase transitions, allowing for investigation into its use in functional magnetic devices.

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Optical investigation of altermagnetism in antiferromagnetic LaFeO₃ thin films

^oKazuya Katsubori¹, Kazunari Ishida¹, Junichi Shiogai^{1,2},
Kohei Ueda^{1,2,3}, Yu Shiratsuchi^{2,3,4}, Kentaro Toyoki^{2,3,4}, Jobu Matsuno^{1,2,3}

¹Graduate School of Science, The University of Osaka, Japan

²Center for Spintronics Research Network, The University of Osaka, Japan

³Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives, The University of Osaka, Japan

⁴Graduate School of Engineering, The University of Osaka

Recently, a novel class of magnetic materials, known as altermagnets, has attracted great attention as a promising platform for spintronics. Although altermagnets exhibit no net magnetization owing to their antiparallel spin order, similar to conventional antiferromagnets, they can host ferromagnet-like phenomena such as spin-split electronic bands and spin pumping owing to the coupling between magnetic order and crystal symmetry. In this work, we focused on the LaFeO₃, which is a 3d transition metal perovskite with an orthorhombic structure attributed to GdFeO₃-type distortion in the bulk. Previous theoretical studies have suggested altermagnetism of LaFeO₃ arising from this distortion [1]. However, transport-based probes are difficult to apply because LaFeO₃ is an insulator. Here, we investigated the magneto-optical Kerr effect (MOKE) of a LaFeO₃/NdGaO₃(110) epitaxial film and observed a characteristic MOKE spectrum consistent with possible altermagnetism in LaFeO₃.

We deposited 26.7 nm thick LFO thin film on NdGaO₃(110) substrate by pulsed laser deposition. The X-ray diffraction pattern shown in Fig. 1(a) confirms the epitaxial growth of a highly crystalline LaFeO₃ thin film. Fig. 1(b) displays magnetic field dependence of its MOKE signal, where clear hysteresis within $\mu_0 H < 0.13$ T is observed. Fig. 1(c) demonstrates photon energy dependence of the saturated Kerr rotation angle of the thin film sample and the substrate. The Kerr spectrum of LaFeO₃ exhibits two sign reversals and a pronounced minimum around $\hbar\omega = 2.85$ eV. This response is consistent with possible altermagnetism in LaFeO₃; the observed spectrum is qualitatively similar to the calculated off-diagonal optical conductivity of LaCrO₃, another proposed altermagnet [2]. However, weak ferromagnetism arising from spin canting can also generate a MOKE signal and may contribute to the observed response. Here we discuss the possible origin of the observed MOKE signal, including contributions from weak ferromagnetism associated with spin canting and the role of structural symmetry.

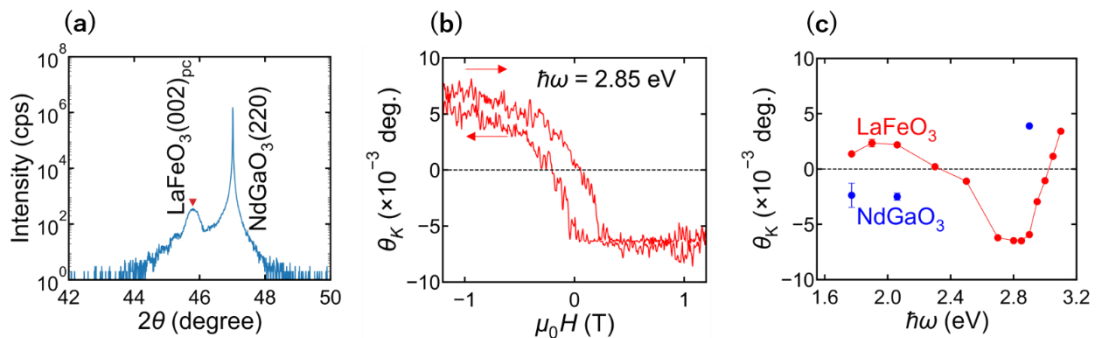


Fig. 1. (a) X-ray diffraction (2θ - ω) pattern of LaFeO₃/NdGaO₃(110) sample. (b) Magnetic-field dependence of the Kerr rotation angle at $\hbar\omega = 2.85$ eV of LaFeO₃/NdGaO₃(110) sample. (c) Photon-energy dependence of the saturated Kerr rotation angle of LaFeO₃ film and NdGaO₃ substrate.

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Magnetic High Entropy Oxides: A New Class of Magnetic Dielectrics

Ryan Thompson¹, R. Capu², C. W. Rischau³, M. R. Cantarino⁴, P. Marsik¹, S. L. Bud'ko⁵, N. Biškup⁶, M. Varela⁶, Y. G. Pashkevich^{1,7}, S. M. Orel⁷, T. Prokscha⁸, A. Suter⁸, J. Zhao⁴, U. Oji⁴, M. Bonura³, P. Bencok⁹, Z. Salman⁸, S. Gariglio³, C. Bernhard¹, S. Sarkar¹

¹ Department of Physics, University of Fribourg, Switzerland

² Department of Physics, West University of Timisoara, Romania

³ Department of Quantum Matter Physics (DQMP), University of Geneva, Geneva, Switzerland

⁴ European Synchrotron Radiation Facility, France

⁵ Ames National Laboratory, Iowa State University, Ames, Iowa, USA

⁶ Departamento de Física de Materiales, Universidad Complutense de Madrid, Madrid, Spain

⁷ O.O. Galkin Donetsk Institute for Physics and Engineering NAS of Ukraine, Kyiv, Ukraine

⁸ PSI Center for Neutron and Muon Sciences CNM, Villigen, Switzerland

⁹ Diamond Light Source, Harwell Science and Innovation Campus, Chilton, Didcot, U.K

We report for the first time the dielectric properties of epitaxial thin films of the magnetic high entropy oxide (HEO) perovskite $\text{Nd}(\text{Cr}_{0.2}\text{Mn}_{0.2}\text{Fe}_{0.2}\text{Co}_{0.2}\text{Ni}_{0.2})\text{O}_3$ (Nd-HEO), which are intriguing as its magnetic properties [1]. Nd-HEO orders magnetically below $T_{\text{mag}} \approx 190$ K, above which the dielectric response reveals a Debye-type frequency dependence with a substantially large zero-frequency dielectric constant of $\epsilon_r' \approx 230$ –250. The dc bias voltage loops are reversible but exhibit three distinct peaks centred at zero and finite positive and negative voltage. In combination with a large dielectric strength, it enables a non-hysteretic tuning of the dielectric response of magnetoelectronic devices with multiple parameters like temperature, electric, and magnetic field. From STEM-EELS and XAS measurements, we provide evidence that the zero-bias peak is governed by the oxygen sublattice while the finite bias peaks originate from cationic dipoles. The maximal response of the latter appears to be shifted to finite bias by a static uncompensated electric field due to a vertical gradient of the oxygen content. Below T_{mag} , this anomalous dielectric response is strongly suppressed and follows the non-magnetic volume fraction as observed from low energy μSR . This happens presumably by magnetostriction that counteracts and freezes the ionic displacements. These findings indicate a unique correlation between configurational entropy, dielectric response, and magnetic properties. This Nd-HEO is equally interesting for fundamental studies of competing electric and magnetic orders in strongly disordered materials.

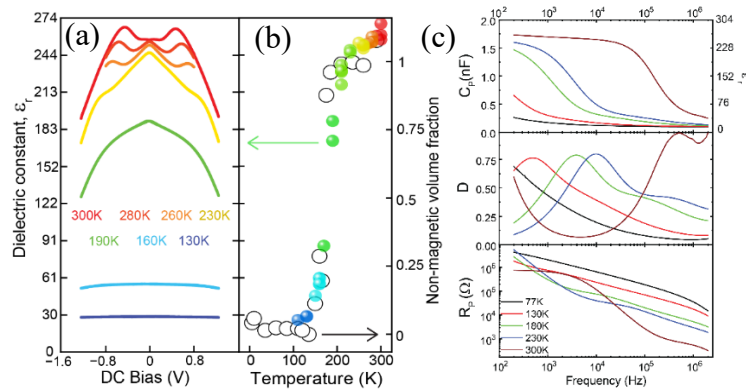


Figure 1: (a) Dielectric response of Nd-HEO as a function of DC bias. (b) ϵ_r' sharply reduces following the non-magnetic volume fraction, indicating coupling between magnetism and dielectric behavior. (c) Frequency and temperature dependence of C_p , ϵ_r' , D , and R_p .

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Spontaneous Voltage and Persistent Current Generation at Cuprate/Manganite Interfaces

Subhrangsu Sarkar^{1*}, Mathias Soulier¹, Shamashis Sengupta², Yurii G. Pashkevich^{1,3}, Roxana Capu⁴, Ryan Thompson¹, Jarji Khmaladze¹, Miguel Monteverde⁵, Louis Dumoulin², Dominik Munzar⁶, Christian Bernhard^{1*}

¹ University of Fribourg, Department of Physics, Fribourg, Switzerland

² Université Paris-Saclay, CNRS/IN2P3, IJCLab, France

³ O. Galkin Donetsk Institute for Physics and Engineering NAS of Ukraine, Ukraine

⁴ West University of Timisoara, Faculty of Physics, Timisoara, Romania

⁵ Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, Orsay, France

⁶ Department of Condensed Matter Physics, Faculty of Science, Masaryk University, Brno, Czech Republic

We present an example of non-reciprocal transport under time-reversal symmetry, which is of great current interest. YBa₂Cu₃O₇(YBCO)/Nd_{0.65}(Ca_{0.7}Sr_{0.3})_{0.35}MnO₃ (NCSMO) multilayers generate spontaneous voltage (SpV) of tens of millivolts over a broad temperature and magnetic field range, which is capable of driving a persistent current through an external load [1]. We provide evidence that this phenomenon occurs as rectification of environmental electromagnetic fluctuations due to an underlying ratchet-type potential presumably originating from the complex domain state of the NCSMO layers which host several nearly degenerate magnetic, electronic and polar orders. Competition between a charge/orbital ordered majority phase with polar moments and a nonpolar ferromagnetic minority phase seems to control the final state. A central role is also played by the adjacent YBCO layers that are too thin (≤ 10 nm) to fully screen the electric fields emanating from the NCSMO layers. Additionally, between 30 K and 80 K, the SPV displays reproducible but hysteretic behaviour under external magnetic field sweep. These multilayers are useful for energy harvesting over broad temperature and magnetic field ranges, and for tunable multifunctional memory devices that are responsive to magnetic fields, electric currents, and electromagnetic radiation.

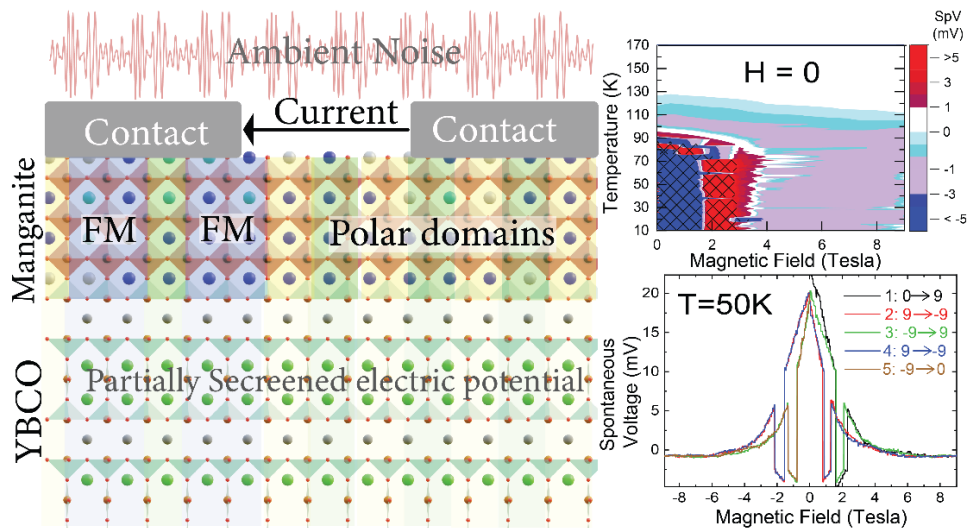


Figure 1: **Sensing of ambient noise by manganite/cuprate heterostructures.** (left) schematic structure of the NCSMO/YBCO interface indicating a phase mixture of polar and FM domains and the YBCO layer under partially screened electric potential subjected to the ambient noise, (right-top) temperature-dependence of SpV at $I=H=0$ and (right-bottom) magnetic-field dependence of SpV at $I=0$ and $T=50$ K.

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Tunable Ru-doped manganite-based synthetic antiferromagnets

L. Guasco^{*1}, J. Schöpf², T. Zander², V. Kabanova^{3,4}, A. Vorobiev^{3,4}, P. H. M. van Loosdrecht², I. Lindfors-Vrejoiu²

¹ Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany

² II. Physics Institute, University of Cologne, 50937 Cologne, Germany

³ Institut Laue-Langevin, 71 avenue des Martyrs, CS 20156, Grenoble Cedex 9, F-38042 France

⁴ Department of Physics and Astronomy, Uppsala University, Box 516, Uppsala, 751 20 Sweden

Synthetic antiferromagnets (SAFs) are at the basis of many spintronics applications such as magnetic tunnel junctions, read heads, and spin-orbit torque devices. SAFs made of correlated oxides introduce additional degrees of freedom and are promising for the realization of more exotic magnetic states beyond the RKKY model. Previous studies confirmed interlayer exchange coupling (IEC) in all-oxide multilayers, mostly combining ferromagnetic manganites with other oxides such as ruthenates [1,2], nickelates [3,4], and even over-doped manganites [5]. All these have a strong in-plane anisotropy, but out-of-plane SAF are preferable due to the absence of stray fields impacting neighbouring bits, crucial for miniaturization. For this end, $\text{La}_{0.3}\text{Sr}_{0.7}\text{Mn}_{1-x}\text{Ru}_x\text{O}_3$ (LSMRO) with strong perpendicular magnetic anisotropy are considered as ferromagnetic layer.

All-oxide SAFs were realized in epitaxial multilayers composed of LSMRO and LaNiO_3 (LNO) spacers [6]. We found a strong IEC with antiparallel alignment of consecutive layers stable up to 0.275 T and at least up to 180 K [7]. Low-field manipulation of the SAF state is enabled by the odd number of ferromagnetic layers, due to the presence of uncompensated magnetic moments (Fig.1a). The interplay between antiferromagnetic IEC, tilted out-of-plane magnetic anisotropy, and structurally favoured domain formation leads to strongly direction-dependent magnetization reversal mechanisms (Fig.1b). Polarized neutron reflectometry is a powerful method to probe the in-plane magnetization components versus external field, offering key insights into the underlying hysteretic behavior and SAF robustness (Fig. 1c,d).

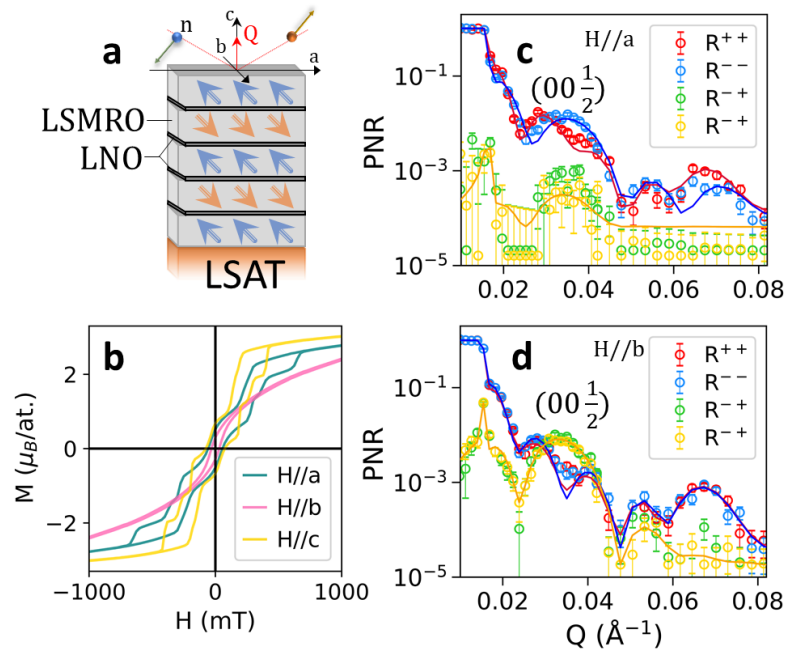


Figure 1: (a) Sketch of the SAF design and experimental geometry, (b) hysteresis loops measured along different directions at 80K; polarized neutron reflectometry (PNR) at 10 mT and 80 K after saturation along the easy in plane axis (c) and hard in plane axis (d).

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Enhanced magnetism in cation-modulated manganite heterostructures

Sangkyun Ryu¹, R. F. Need², Brian Kirby³, Tae-Yeol Jeon⁴, Younghak Kim⁴, Jin H. Cho⁵,
Sungkyun Park^{1,6} and Hyoungjeen Jeon^{1,6}

¹*Research Center for Dielectric and Advanced Matter Physics, Pusan National University, Busan 46241, Korea*

²*Qualcomm Institute, University of California San Diego, La Jolla, CA 92093 USA*

³*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA*

⁴*Pohang Accelerator Laboratory, POSTECH, Pohang 37673, Korea*

⁵*Department of Physics Education, Pusan National University, Busan 46241, Korea*

⁶*Department of Physics, Pusan National University, Busan 46241, Korea Author*

Transition-metal oxides exhibit a variety of physical properties that can be tuned by external stimuli such as electric and magnetic fields, mechanical stress, and interfacial interaction. Among these, interfacial interactions are well known, including charge transfer, polarization enhancement, and the superconducting transition.[1] These interfacial interactions are effective ways to improve and tune magnetic properties at the manganite.[2] Usual interfacial interaction studies have been focused on cases with large valence contrasts. It remains unclear how this applies to the manganite superlattice with small differences in valence state. In this presentation, we grow NSMO SLs with a small cation stoichiometry. To deposit the $\text{Nd}_{1-x}\text{Sr}_x\text{MnO}_3$ (NSMO) SLs, an $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ target was used, and the cation ratio was controlled by the oxygen partial pressure during one-pot synthesis.[3] Using X-ray and neutron methods, we observed structural properties and the formation of NSMO SLs. In addition, the magnetic depth profile was obtained by polarized neutron reflectivity. The magnetic properties of the NSMO SLs were observed by SQUID magnetometry and compared to NSMO single-layer films. To observe changes in the valency of manganese, we use X-ray absorption spectroscopy. These results help in understanding the interaction between magnetic materials and improving their properties.

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Giant Magnetization Enhancement in an Ultrathin Orthoferrite

Jin Young Oh¹, Yaolong Xing², Seung Gyo Jeong¹, Sehwan Song³, Woo-suk Noh⁴, Valeria Lauter⁵, Seongjae Choi⁶, Seyoung Kwon³, Jaeha Choi³, Si Won Kim¹, Hong Hyun Jeon¹, Heemin Lee⁷, Cheng-Tai Kuo⁷, Jun-Sik Lee⁷, Heung-Sik Kim², Sungkyun Park³, Sang Ho Oh², Woo Seok Choi^{1*}

¹ Department of Physics, Sungkyunkwan University, Suwon, South Korea

² Department of Energy Engineering, Korea Institute of Energy Technology, Naju, South Korea

³ Department of Physics, Pusan National University, Busan, South Korea

⁴ MPPC-CPM, Max Planck POSTECH Korea Research Initiative, Pohang, South Korea

⁵ Spallation Neutron Source, Oak Ridge National Laboratory, TN, USA

⁶ Department of Semiconductor Physics, Kangwon National University, Kangwon, South Korea

⁷ Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, CA, USA

Low dimensional magnetic materials have received significant attention due to their high scalability and energy efficiency for future electromagnetic applications. Despite these advantages, low dimensional magnetism at finite temperature is subject to Mermin-Wagner fluctuation, which renders the realization highly challenging. Here, we present microscopic structural modulation in epitaxial ultrathin film of an orthoferrite to achieve emergent magnetic response at room temperature. ErFeO₃ epitaxial thin films with atomically precise thickness control demonstrate that the saturation magnetization is as high as 3.7 μ_B /f.u. for the 2-nm-thick epitaxial film, which is unimaginable for bulk ErFeO₃ which is antiferromagnetic. The characteristic domain structure resultant from epitaxial strain and orbital reconstruction at the surface, both arising from the thin film geometry, have been proposed as the underlying mechanism of the giant magnetization enhancement. Our results imply that the magnetic ordering can indeed be realized in two dimensional limit by deliberate structural control of orthoferrite systems.

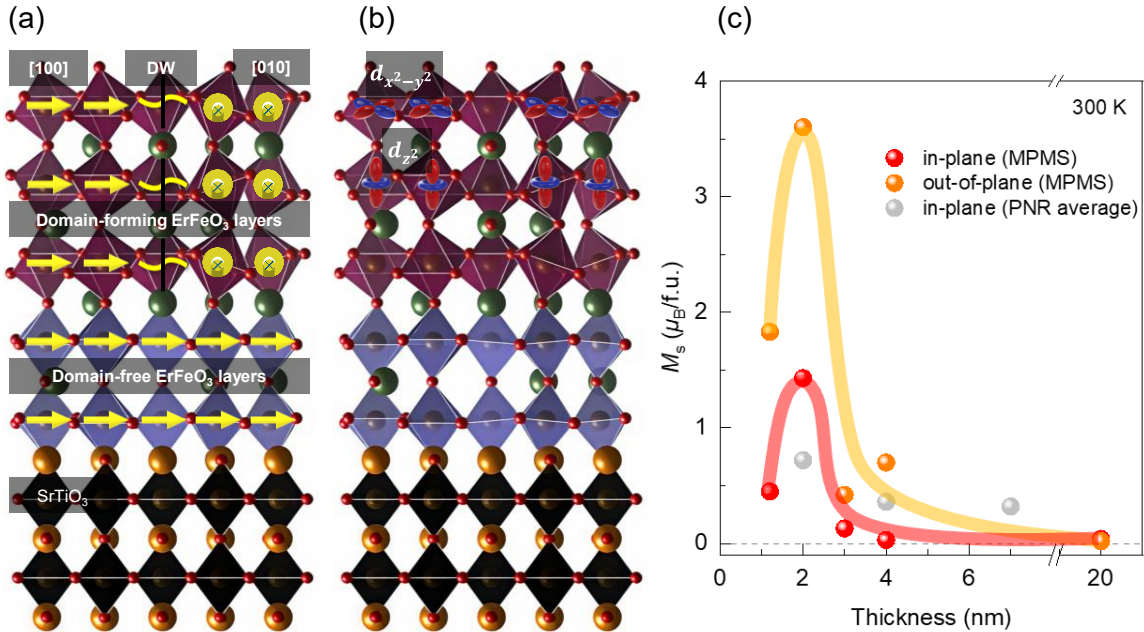


Figure 1: Schematic diagrams of (a) structural domain effect, which may block the propagation of exchange interaction and (b) surface orbital reconstruction effect, which weakens antiferromagnetic exchange interaction in ErFeO₃ ultrathin films. (c) Magnetization as a function of film thickness obtained from various measurements including magnetic property measurement system and polarized neutron reflectometry at 300 K.

Thin Film Synthesis of a New Rocksalt-Type Rare-Earth Monoxide TmO with Ferromagnetism and Multicarrier Conduction

Hidetatsu Yoshimura¹, Masamichi Negishi¹, Satoshi Sasaki¹, Daichi Oka², Daisuke Shiga^{3,4}, Hiroshi Kumigashira^{3,4} and Tomoteru Fukumura^{1,5,6}

¹ Department of Chemistry, Graduate School of Science, Tohoku University, Sendai, Japan

² Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University, Tokyo, Japan

³ Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai, Japan

⁴ Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, Japan

⁵ Advanced Institute for Materials Research (WPI-AIMR), Tohoku University, Sendai, Japan

⁶ Center for Science and Innovation in Spintronics, Tohoku University, Sendai, Japan

Recently, most of metastable rocksalt-type rare-earth monoxides (REOs) have been synthesized with thin film epitaxy, unveiling their electronic and magnetic properties [1]. Thulium monoxide (TmO) is the last REO to be synthesized. Although the other REOs have often been obtained in a form of single phase epitaxial thin film, TmO phase is chemically unstable due to its lower formation energy than other REOs [2]. In this study, we synthesized rocksalt-type TmO and measured electronic and magnetic properties for the first time.

TmO thin films were deposited on CaF₂(001) substrates from a Tm₂O₃ target by the pulsed laser deposition method. Epitaxial growth of rocksalt-type TmO was confirmed in addition to Tm₂O₃ phase, which is insulating and paramagnetic, with X-ray diffraction (Fig. 1). X-ray photoemission and absorption spectroscopy revealed a 4f¹²5d¹ electronic state and a metallic Fermi edge of TmO. TmO thin film was ferromagnetic below 56 K (Fig. 2). This Curie temperature (T_C) was consistent with its chemical trend of other heavy REOs, where the T_C monotonically increases with the de Gennes factor [3]. The Hall effect measurements showed a transition from negative to positive Hall coefficient with decreasing temperature (Fig. 3), suggesting multicarrier conduction. The anomalous Hall effect was observed below the T_C , and the hysteresis loops were in an unusual direction (Fig. 3). This result suggests multiple components of anomalous and/or nonlinear Hall effect as was observed in ErO [4].

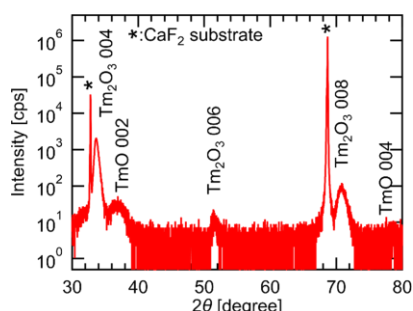


Figure 1: X-ray diffraction 2θ - ω pattern for TmO thin film.

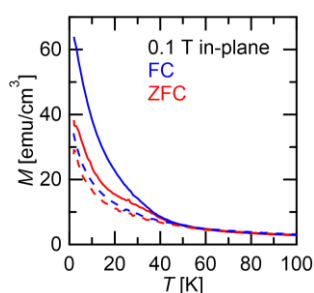


Figure 2: Temperature dependence of magnetization for TmO thin film (solid lines) and referential Tm₂O₃ film (dashed lines).

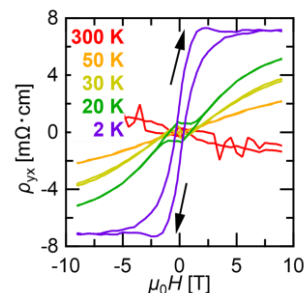


Figure 3: Magnetic field dependence of Hall resistivity for TmO thin film.

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Emergent Magnetic and Electrical Transport Properties in 4d/5d Transition-Metal-Substituted LSMO

Kenichi Kaminaga¹, Gaku Sato¹, Yuto Abiko¹, Kenji Itaka², Tetsuro Ueno³, Yoshiyuki Ohtsubo³,
Takayasu Hanashima⁴, Kazuhiro Akutsu-Suyama⁴, Shingo Maruyama^{1,5}, Hiroyuki Aoki⁶,
and Yuji Matsumoto¹

¹ Tohoku University

² Hirosaki University

³ National Institutes for Quantum Science and Technology (QST)

⁴ Comprehensive Research Organization for Science and Society (CROSS)

⁵ Shizuoka University

⁶ Japan Atomic Energy Agency (JAEA)

(La,Sr)MnO₃ (LSMO) is a leading candidate for oxide spintronics and magnetic memory applications owing to its high spin polarization and robust room-temperature ferromagnetism, whereas its extremely low coercivity remains a drawback. Partial substitution of Mn atoms with 4d/5d heavy transition metal atoms offers a route to controlling magnetic anisotropy, spin relaxation, and electrical transport through strong spin–orbit coupling. Among them is 4d-element-substituted Ru:LSMO, where the Ru substitution enhances coercivity (H_c), but typically lowers saturation magnetization (M_s) and Curie temperature (T_C) as long as the substitution is uniform [1].

To overcome this trade-off, we extended the concept of graded magnetism, originally developed in metallic ferromagnets films [2], to complex-oxide ones. Ru:LSMO epitaxial films with linearly graded Ru composition were fabricated by pulsed laser deposition [3]. Macroscopic magnetometry revealed a 7–10-fold enhancement in H_c while preserving M_s and effective T_C comparable to those of pristine LSMO, demonstrating that the concept of graded magnetism can decouple competing magnetic parameters through continuous exchange coupling between Ru-rich and Ru-poor regions [4].

To elucidate the microscopic origin, we employed a combined approach of depth-resolved polarized neutron reflectometry (PNR) at BL17 (J-PARC MLF) and Mn $L_{2,3}$ -edge fluorescence-yield X-ray magnetic circular dichroism (FY-XMCD) at BL13U (NanoTerasu). PNR revealed that, despite reduced magnetization in the interfacial and near-surface dead layers, the intervening region exhibited locally enhanced magnetization exceeding that of the corresponding uniformly substituted film. FY-XMCD confirmed that this reinforcement originated from the Mn spin sublattice itself.

These findings establish a new pathway toward advanced oxide spintronics through substitutional-dopant depth-profile engineering, which provides an additional degree of freedom for overcoming trade-offs among H_c , M_s , and T_C . In the presentation, as a brief extension to 5d-element-substituted LSMO, we will also show anomalous zero-resistance-like behavior observed in four-terminal transport measurements of Ir-substituted LSMO.

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Magnetoresistance governs apparent magnetic-field enhancements in water electrolysis

Vadim Ratovskii¹, Emma van der Minne¹, Jelle R. H. Ruiters¹, Iris C. G. van den Bosch¹, Michael Verhage², Daan H. Wielens¹, Gertjan Koster¹, Christoph Baeumer^{1,3}

¹ MESA+ Institute, Faculty of Science and Technology, University of Twente, Enschede, The Netherlands

² Materials to Optoelectronic Devices (M2D), Departments of Applied Physics and Chemical Engineering and Chemistry, Eindhoven University of Technology, Eindhoven, The Netherlands.

³ Peter Gruenberg Institute 7, Forschungszentrum Juelich GmbH, Juelich, Germany.

The transition to green energy relies heavily on efficient energy storage and utilization, with hydrogen offering a sustainable solution. While water electrolysis provides a sustainable route for hydrogen generation, the oxygen evolution reaction (OER) remains a kinetic bottleneck due to its sluggish four-electron transfer process kinetics. Although transition metal oxides are promising OER catalysts because of their earth abundance and high activity, additional research is needed to further improve their performance. Recently, it has been reported that spin-ordered materials under magnetic fields can enhance OER catalytic efficiency [1], offering a route toward a more energy-efficient green hydrogen economy. The exact origin of this enhancement, however, remains unclear and requires a novel approach to understand the nature of the effect.

Epitaxial films as model systems are particularly attractive to study the fundamentals of water electrolysis [2], as we also outlined in our recent roadmap for the emerging field of spin-enhanced electrocatalysis [3]. In this work, we use epitaxial room-temperature ferrimagnetic CoFe_2O_4 (CFO) - based heterostructures to reveal the origin of the magnetic enhancement effect [4]. The samples were prepared by pulsed laser deposition and characterized in terms of surface morphology, magnetic properties, crystallinity, and oxidation state. By systematically varying the conductive buffer layer beneath the insulating CFO surface between paramagnetic and ferromagnetic states (Figure 1a), we disentangle the respective contributions of surface magnetic domain alignment and magnetoresistance (MR) to the observed OER current enhancement (Figure 2b). The enhancement persists with increasing magnetic field even after the system reaches a monodomain state, indicating no correlation with domain wall removal, previously proposed as the dominant mechanism behind this effect [5]. Our results show that the OER current increase directly correlates with the MR effect, identifying it as the primary origin of magnetic-field-induced OER enhancement.

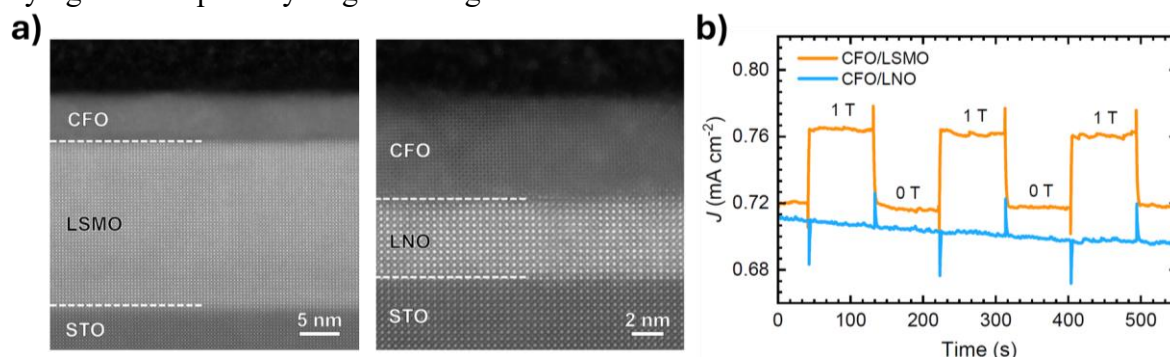


Figure 1: a) TEM images of CFO/LSMO and CFO/LNO based heterostructures and b) their electrochemical performance in 0 T and 1 T magnetic fields.

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Evolution of the Spin Cycloid in BiFeO₃ Thin Films with Magnetic Field and Temperature

Manu Mohan¹, Hamish Mountfort¹, Daniel Sando¹

¹MacDiarmid Institute for Advanced Materials and Nanotechnology, School of Physical and Chemical Sciences, University of Canterbury, Christchurch, New Zealand

BiFeO₃ (BFO) is one of the most heavily investigated multiferroic materials due to the interesting coexistence of ferroelectric and antiferromagnetic orders well above room temperature. This robust multi-functionality makes it the best candidate for magnetoelectric coupling, electrically controlled magnetism, and spin-based devices. The application potential of BFO is highly influenced by the presence of cycloid spin structure, which arises due to the balance of exchange interactions, magnetic anisotropy and Dzyaloshinskii-Moriya Interaction [1]. The cycloid modulations reduce the net weak magnetic moment but unlocking complex magnetic dynamics and electrically coupled spin excitations. Hence, understanding and controlling this modulation is a central challenge for future magnetic and spintronic architectures [2].

Although the response of the cycloid to external stimuli has been extensively explored in BFO single crystals [3] and actively investigated in thin films at low temperatures near 150 K [4], a comprehensive picture extending to room temperature remains unexplored. The strain induced by the substrate alters the balance between magnetic interactions, making thin-film architecture an exceptionally attractive playground for practical device applications. However, a systematic understanding of the cycloidal structure at room temperature under magnetic field, as well as its thermal and temporal evolution, is not yet well established in BFO films.

This work uses neutron diffraction to map the evolution of the cycloidal modulation under external magnetic field and temperature in a (101)_{pc} oriented BFO thin film. The variation in cycloid period is studied as a function of magnetic field (0 to 10 T) applied along the [10 $\bar{1}$] direction, orthogonal to polarisation and cycloid propagation, at different temperatures. These results, particularly the presence of a hysteresis effect, strongly suggest that the cycloidal to pseudo-collinear G-AFM transition is first order in nature. In addition, thermal and temporal stability of cycloids are extracted from neutron maps recorded during a temperature sweep and at different times. These works provide a comprehensive insight into the evolution of complex spin structures under the influence of magnetic field, temperature and time, facilitating the controlled manipulation of cycloids for future functional oxide devices.

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Growth control during pulsed laser deposition to tune the electrochemical behavior of epitaxial $\text{Li}_4\text{Ti}_5\text{O}_{12}$ spinel thin films

Seung Heon Han¹, Guus Rijnders¹, Mark Huijben¹

¹ MESA+ Institute for Nanotechnology, University of Twente, 7500 AE Enschede, The Netherlands

A fundamental understanding of solid-state thin-film batteries is essential for realizing a highly interconnected society powered by next-generation microelectronic systems, including Internet of Things (IoT) sensors, medical implants, and wearable devices [1,2]. However, developing a comprehensive understanding of such systems remains challenging because their behavior is often complicated by the various chemical additives commonly employed in conventional batteries. Epitaxial growth addresses the challenge by providing simplified thin-film model systems for investigating the intrinsic electrochemical properties, including specific capacity, rate capability, and cycling stability [3]. In this study, epitaxial lithium titanate spinel oxide thin films ($\text{Li}_4\text{Ti}_5\text{O}_{12}$, LTO) were realized by pulsed laser deposition (PLD) to elucidate how laser ablation conditions influence thin-film growth and electrochemical Li-ion storage behavior. The laser repetition rate and spot size on the target were systematically varied while all other deposition conditions were maintained constant. Structural, morphological, and electrochemical analyses demonstrated that these parameters strongly affected the growth behavior, crystallographic characteristics, surface morphology, and electrochemical response of the LTO thin films. These findings demonstrate that laser ablation parameters can substantially influence the structure and the corresponding electrochemical behavior of epitaxial LTO thin-film electrodes, highlighting the importance of carefully controlling these parameters in PLD studies of functional oxide materials containing volatile elements such as lithium.

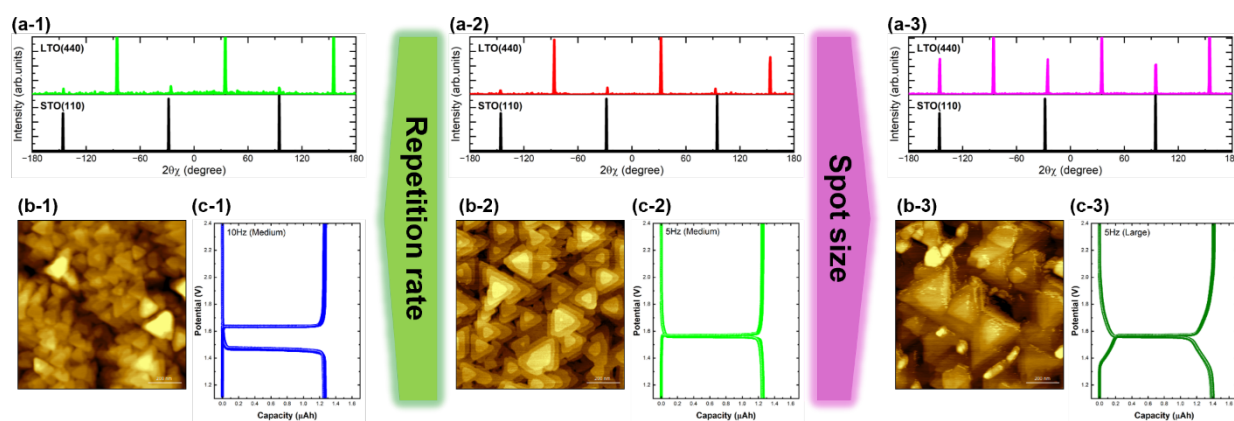


Figure. (a) Phi scans of the LTO (440) and STO (110) reflections measured for epitaxial LTO thin films. (b) AFM surface morphologies and (c) galvanostatic charge-discharge profiles of the epitaxial LTO films. Samples 1, 2, and 3 correspond to films deposited at 10Hz with a medium spot size, 5Hz with a medium spot size, and 5Hz with a large spot size, respectively.

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Towards Tunable Non-Collinear Magnetism in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ Superlattices

Jan Klimek^{1,2}, Gideok Kim¹, Laura Guasco¹, Michael Neumann¹, Adam Dubroka²,
Bernhard Keimer¹

¹ *Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany*

² *Department of Condensed Matter Physics, Faculty of Science,
Masaryk University, Brno, Czech Republic*

Correlated oxide heterostructures provide a powerful route to engineer emergent magnetic states through interfacial exchange coupling, charge transfer, and orbital reconstruction. In $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO), Sr doping enables tuning between ferromagnetic (FM) and antiferromagnetic (AF) order, allowing local control of competing interactions. By combining layers with different doping levels, LSMO superlattices form a natural platform for complex magnetic configurations. In our previous work, this approach led to the observation of a doubled non-collinear “magnetic fan” structure under selected conditions [1].

Here we study a systematic series of 18 LSMO homojunction superlattices grown by ozone-assisted molecular beam epitaxy, varying both Sr doping and the thickness of FM and AF layers. Structural quality and periodicity were determined by X-ray reflectometry and diffraction.

Magnetic measurements reveal a distinct region with non-trivial behavior. Selected superlattices exhibit sub-loop features in the hysteresis below ~ 150 K (Fig. 1), evolving systematically across the series and indicating competing interlayer exchange. Magnetotransport shows corresponding low-field anomalies with comparable field scales, suggesting a common origin and strong sensitivity to the magnetic configuration.

These combined signatures strongly indicate that fan-like non-collinear magnetic states are realized in this region. The systematic dataset provides a basis for quantitative understanding of interlayer exchange coupling and for developing models that describe the magnetic fan structure, enabling targeted design of oxide superlattices with tunable non-collinear magnetism for device applications.

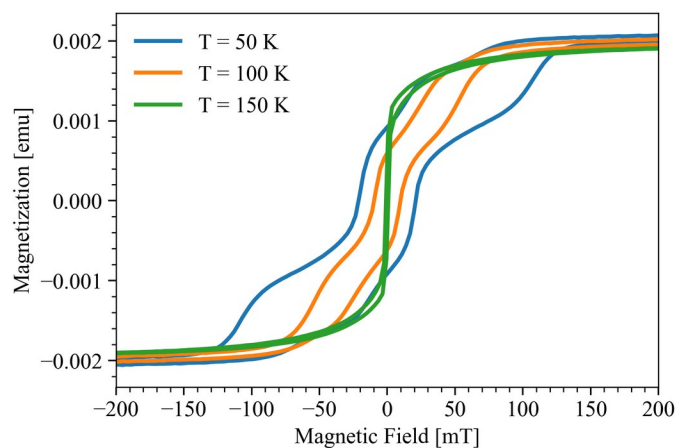


Figure 1: Hysteresis loops of superlattice $10x[9x(\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3)5x(\text{La}_{0.3}\text{Sr}_{0.7}\text{MnO}_3)]$ exhibiting onset of complex hysteresis behavior under 150 K.

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Single-Phase RockSalt-Type Gadolinium Monoxide Epitaxial Thin Films with a Large-Thickness-Dependent Magnetization

Satoshi Sasaki¹, Masamichi Negishi¹, Daisuke Shiga^{2,3}, Hiroshi Kumigashira^{2,3}, Tomoteru Fukumura^{1,4,5}

¹ Department of Chemistry, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan

² Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan

³ Photon Factory, Institute of Materials structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba 305-0801, Japan

⁴ WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

⁵ Center for Science and Innovation in Spintronics, Organization for Advanced Studies, Tohoku University, Sendai 980-8577, Japan

Stable binary rare earth (*RE*) oxides are highly insulating and mostly nonmagnetic sesquioxides (RE_2O_3) with trivalent *RE* ions. Recently, rocksalt-type rare earth monoxides (*RE*O) with unusual divalent *RE* ions were synthesized via nonequilibrium thin film epitaxy. In contrast to RE_2O_3 , *RE*O exhibited much higher electrical conductivity owing to the $[Xe]4f^n5d^1$ electron configuration of RE^{2+} ions containing itinerant 5d electrons, as well as a variety of magnetism [1]. Among them, light *RE*O (*RE* = La–Sm) possessed paramagnetism or low Curie temperature (T_C) ferromagnetism, whereas several heavy *RE*O (*RE* = Gd, Tb, Ho) possessed significantly higher T_C than light *RE*O. In particular, room-temperature ferromagnetism of GdO is attractive for magnetic and spintronic applications. However, the nonnegligible Gd_2O_3 , resulting from chemical instability and large lattice mismatch of GdO, obscured the intrinsic electronic and magnetic properties [2,3].

In this study, single-phase GdO epitaxial thin films were successfully grown by introducing a lattice-matching (Ca,Sr)O buffer layer with a tunable lattice constant [4]. Synchrotron X-ray spectroscopies confirmed the formally divalent Gd ions with the $[Xe]4f^75d^1$ electron configuration, which is consistent with the metallic conduction showing an order-of-magnitude higher conductivity than impurity-containing GdO thin films (Figure 1). Magnetization measurements confirmed the T_C of 308 K even for 5 nm-thick film. In addition, the non-monotonic thickness dependence of the magnetization was observed (Figure 2), suggesting a non-simple spin texture as a result of competing ferromagnetic and antiferromagnetic exchange interactions.

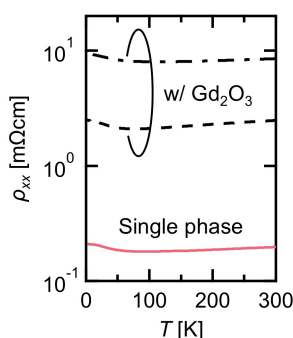


Figure 1. Temperature dependence of the resistivity for single-phase and impurity-containing GdO films.

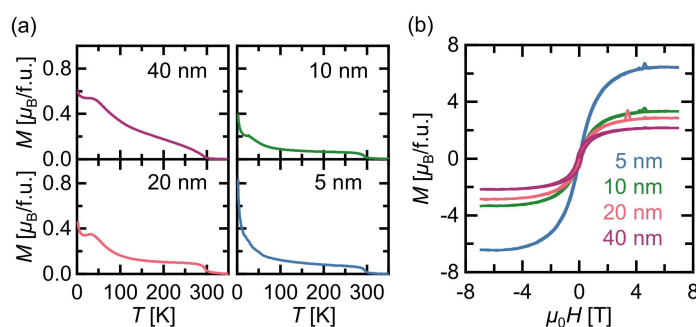


Figure 2. (a) Temperature and (b) magnetic field dependence of the magnetization for single-phase GdO films with different thickness.

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Towards Efficient Spin-Charge Interconversion in γ -Al₂O₃/KTaO₃ Two-Dimensional Electron Gases

Anshu Gupta¹, Amit Chanda¹, Felix Trier¹

1 – Department of Energy conversion and storage, Technical University of Denmark, Denmark.

Abstract

Two-dimensional electron gases (2DEGs) at oxide interfaces provide a versatile platform for exploring emergent electronic and spintronic phenomena.¹ Among them, KTaO₃-based heterostructures have recently attracted considerable interest due to their strong intrinsic spin–orbit coupling arising from the heavy Ta 5d orbitals, offering new opportunities for efficient spin-charge interconversion.²

Here, we report the formation and magneto-transport characterization of a 2DEG at the γ -Al₂O₃/KTaO₃ (GAO/KTO) interface grown by pulsed laser deposition at 350 °C. Electrical transport measurements reveal metallic conduction down to 2 K, while Hall measurements confirm a high sheet carrier density of approximately $4\text{--}9 \times 10^{13} \text{ cm}^{-2}$, and mobility of $\sim 380 \text{ cm}^2/\text{Vs}$ at 2 K. Low-temperature out-of-plane magneto-conductance exhibits robust weak anti-localization behavior below 15K, indicating strong interfacial spin-orbit coupling persisting up to higher temperature than in GAO/STO based 2DEGs.

Therefore, compared to the extensively studied γ -Al₂O₃/SrTiO₃ interface,³ the GAO/KTO heterostructure offers enhanced spin-orbit interaction, making it an attractive platform for spin-orbitronic functionalities. Motivated by these observations, we investigate charge-to-spin conversion at the interface using bilinear magnetoelectric resistance (BMER) measurements, a sensitive probe of spin-momentum-locked transport and Rashba-type spin splitting.

The combination of a high-density 2DEG and strong spin–orbit coupling positions the GAO/KTO interface as a promising candidate for studying spin-charge interconversion and developing future oxide-based spintronic devices.

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Fabrication and physical properties of MgTi_2O_4 spinel oxide thin films

K. Sato^{1,2}, H. Sato², M. Nakano¹, Y. Okada²

¹*Coll. of Eng., Shibaura Inst. of Technol*

²*Okinawa Inst. of Sci. and Tech. Grad. Univ*

Spinel oxide MgTi_2O_4 has attracted significant interest as a platform for diverse quantum phenomena derived from strong electron correlations and geometric frustration inherent to its unique crystal structure. In particular, the (111) plane forms characteristic sublattices, such as a Kagome lattice, raising expectations for thin-film fabrication to explore unknown quantum phases. However, synthesizing stoichiometric MgTi_2O_4 thin films is highly challenging because the titanium valence must be composed entirely of Ti^{3+} , which is thermodynamically unstable in the atmosphere. Due to this synthesis difficulty, previous studies on the epitaxial growth of MgTi_2O_4 thin films have been strictly limited [1, 2].

In this study, we attempted to fabricate MgTi_2O_4 thin films using pulsed laser deposition (PLD). As a result of systematic structural evaluation by varying the deposition parameters, extremely smooth two-dimensional surface growth accompanied by clear streak patterns in *in-situ* RHEED was observed under specific conditions. Furthermore, a distinct reflection from the (111) plane of MgTi_2O_4 was obtained in XRD measurements, confirming the formation of the target spinel phase on a bulk scale. In addition, XPS measurements of the fabricated thin films revealed clear peaks originating from the Ti^{3+} state—which is typically easily oxidized—thereby chemically demonstrating the successful fabrication of the highly unstable MgTi_2O_4 phase.

In this presentation, we will report on the optimal growth conditions for MgTi_2O_4 thin films. We will also discuss in detail the influence of the deposition process on the Ti valence state and the resulting physical properties.

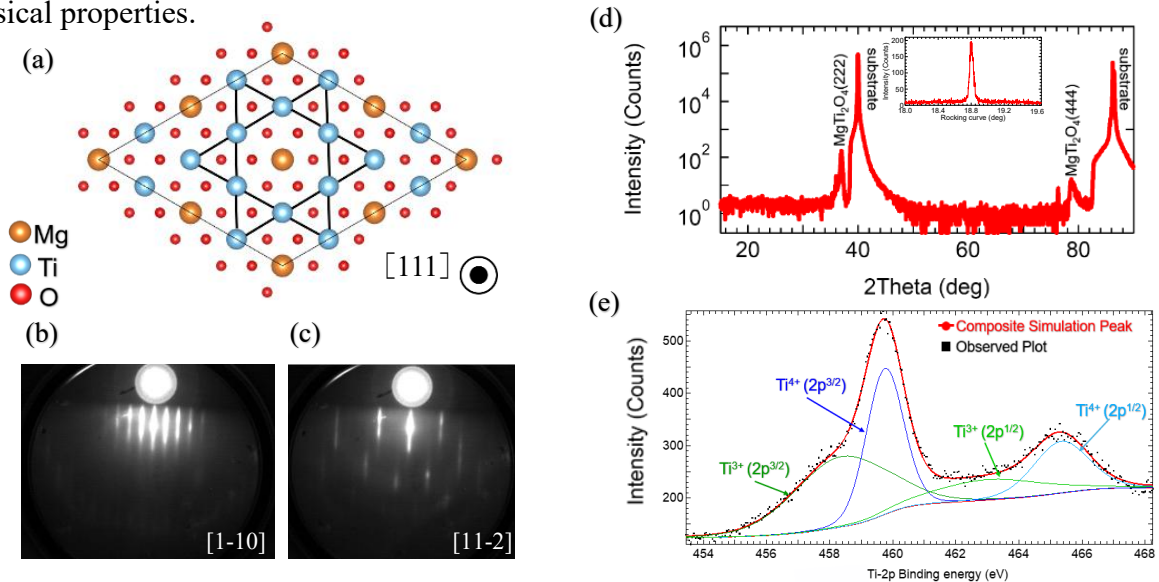


Figure 1: (a) Crystal structure of MgTi_2O_4 (111) with a Kagome lattice (b),(c) *In-situ* RHEED patterns of the fabricated MgTi_2O_4 thin film. (d) XRD 2θ - ω scan of the MgTi_2O_4 thin film. The inset shows the rocking curve of the (222) peak. (e) Ti 2p XPS spectrum of the fabricated thin film

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Interplay of Spin–Orbit Coupling and e-e Coulomb Interactions in BaSnO₃/LaInO₃ Oxide Interfaces Revealed by Magnetotransport

Soumyakanta Panda¹, Georg Hoffmann², Roman Engel-Herbert², Manuel Bibes¹, and Agnès Barthélémy¹

¹ *Laboratoire Albert Fert, CNRS, Thales, Université Paris-Saclay, Palaiseau, France*

² *Paul Drude Institute for Solid State Electronics, Hausvogteiplatz 5-7, 10117 Berlin, Germany*

Spin-orbit interaction (SOI) in two-dimensional electron gases (2DEGs) formed at the surfaces and interfaces of perovskite transition-metal oxides has attracted considerable interest due to its influence on electronic transport properties [1]. Although several experimental studies have reported signatures of SOI in these systems, its role in low-temperature quantum transport remains under debate. In particular, the observation of negative magnetoconductance, often interpreted as a manifestation of weak antilocalization (WAL), does not by itself distinguish between contributions arising from SOI, electron-electron interactions (EEI), or their combined effects [2]. In this work, we present a comprehensive magneto transport investigation of the BaSnO₃/LaInO₃ oxide heterointerface and establish a framework for distinguishing the individual contributions of SOI and EEI at low-temperature transport. By analysing the quantum corrections to sheet conductance in the two-dimensional transport regime, we find that SOI gives rise to a pronounced WAL contribution in the weak magnetic field limit at 2 K. However, for magnetic fields exceeding approximately 100 Oe, electron–electron interaction (EEI) effects become dominant and partially obscure the SOI-induced WAL signature. This behaviour suggests a substantial orbital contribution to the electron transport mechanism.

We report the first observation of a Rashba type surface state in BaSnO₃/LaInO₃ oxide interfaces, with an estimated Rashba coefficient of approximately $10 \text{ meV} - \text{\AA}$. Analysis based on the WL/WAL framework yields spin orbit and inelastic scattering fields of about 4 T and 0.35 T, respectively, at 2 K. A crossover between these scattering strengths is observed near 12 K, indicating a transition from a WAL-dominated transport regime at low temperatures to a WL-dominated regime at higher temperatures. These findings provide new insight into the interplay between SOI and EEI and advances the understanding of quantum interference effects in correlated oxide systems. The observation of a Rashba surface state in this new oxide heterostructure highlights its potential as an attractive platform for studying spin–charge interconversion phenomena and realizing next-generation Rashba-based spintronic devices.

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Spin current generation arising from an epitaxial 4d MoO₂ film

Daigo Takafuji¹, Kohei Ueda^{1,2,3}, Junichi Shiogai^{1,3}, Takanori Kida⁴, Masayuki Hagiwara⁴,
Jobu Matsuno^{1,2,3}

¹*Department of Physics, Graduate School of Science, The University of Osaka, Japan*

²*Center for Spintronics Research Network, The University of Osaka, Japan*

³*Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives, The University of Osaka, Japan*

⁴*Center for Advanced High Magnetic Field Science, The University of Osaka, Japan*

Spin current generation has attracted much attention for magnetization control with low power consumption, enabling next-generation magnetic device application. The spin Hall effect has been known as one of the crucial mechanisms for charge to spin current conversion. It is predicted that the conversion efficiency of transition metal oxides (TMOs) depends on the spin–orbit coupling strength and d -electron number [1]. Although investigations of spin current generation have recently expanded to 5d TMOs [2,3], a comprehensive understanding remains to be elucidated owing to the difficulty of film fabrication. In this talk, we present the room-temperature spin current generation in the 4d rutile-type MoO₂ ($4d^2$) films, grown on an Al₂O₃(0001) substrate by using reactive sputtering.

The thin-film growth conditions were optimized based on structural and electrical conductivity characterization. The optimal MoO₂ film exhibits (010)-oriented monoclinically distorted rutile structure and metallic conductivity, characterized by X-ray diffraction (XRD), X-ray photoelectron spectroscopy, and transport measurements. Figure 1(a) shows the XRD pattern of the MoO₂ film under the optimized condition. The conversion efficiency is evaluated by second-harmonic Hall measurements in the Hall-bar devices fabricated from ferromagnetic Co₂₀Fe₆₀B₂₀ and MoO₂ bilayers. Figures 1(b) and 1(c) show the external field dependence of dampinglike (DL) contributions extracted from second-harmonic Hall resistance $R_H^{2\omega}$, yielding the conversion efficiency of +0.044 for MoO₂ and −0.0041 for control sample Mo. The efficiency of MoO₂ is an order of magnitude larger than that of Mo. The positive sign of the conversion efficiency for MoO₂ in contrast to the negative sign for Mo is consistent with the theoretical study of spin Hall effect based on 4d oxides [1]. In this talk, we will discuss the spin current physics by comparing the conversion efficiencies of MoO₂ ($4d^2$), WO₂ ($5d^2$), and IrO₂ ($5d^5$), which differ in spin–orbit coupling strength and d -electron number.

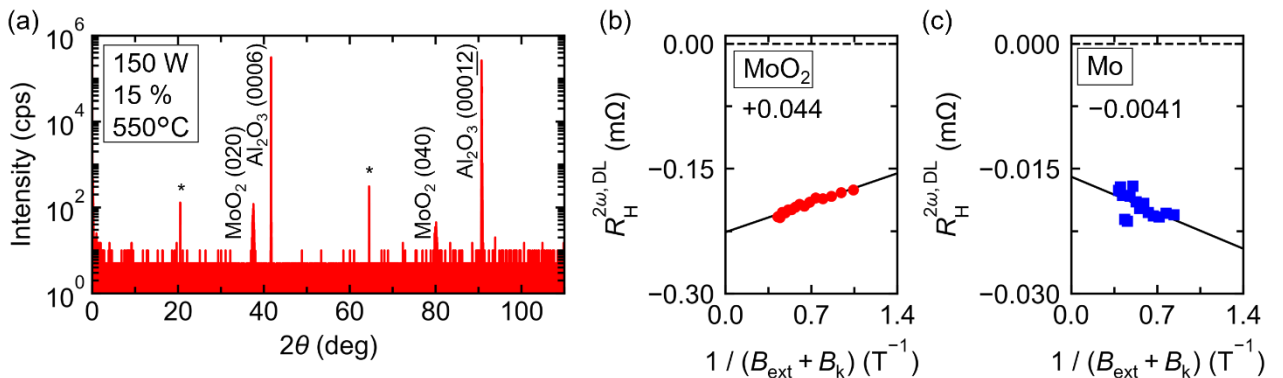


Figure 1: (a) X-ray diffraction pattern of an optimal MoO₂ film grown on an Al₂O₃(0001) substrate under sputtering conditions with a power of 150 W, oxygen partial pressure of 15%, and substrate temperature of 550°C. (b,c) Dampinglike (DL) contributions extracted from second-harmonic Hall resistance for MoO₂ (b) and Mo (c), respectively.

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Spin current generation arising from unique electronic structure of 5d CaIrO_3

Kohei Ueda^{1,2,3}, Ayame Ochi¹, Junichi Shiogai^{1,3}, Jobu Matsuno^{1,2,3}

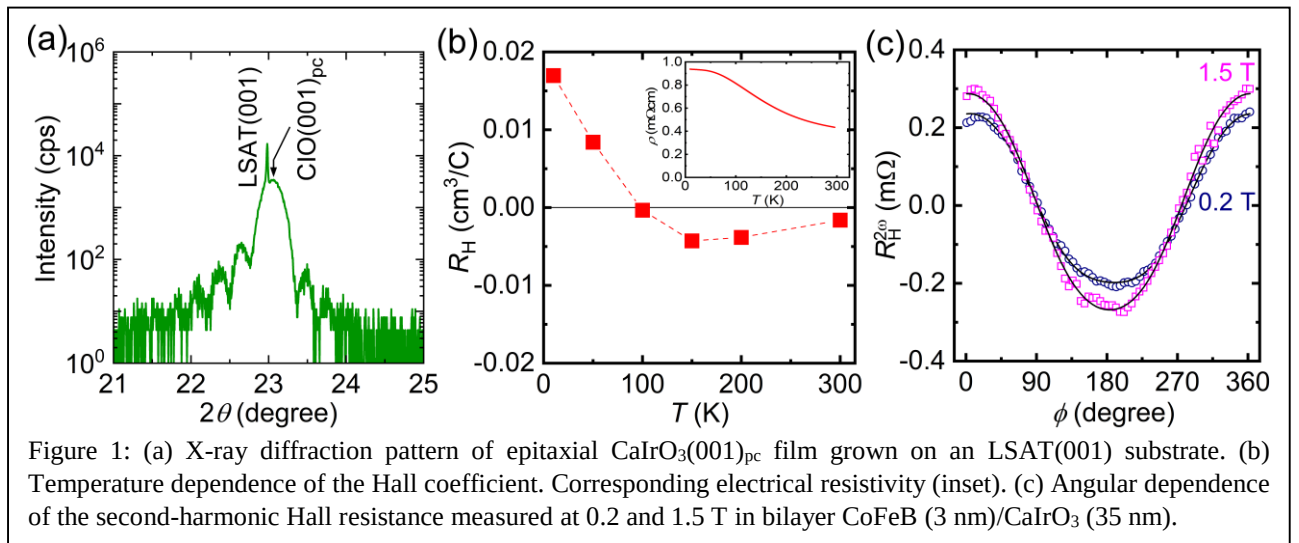
¹*Department of Physics, Graduate School of Science, The University of Osaka, Japan*

²*Center for Spintronics Research Network, The University of Osaka, Japan*

³*Spintronics Research Network Division, Institute for Open and Transdisciplinary Research Initiatives, The University of Osaka, Japan*

Charge to spin current conversion stemming from strong spin-orbit coupling (SOC) is an underlying technology for manipulating the magnetization of magnetic devices with low power consumption. Recently, the 5d transition-metal oxides have provided an intriguing platform for generating an efficient spin current generation due to Fermi level dominated by 5d electrons with strong SOC; perovskite-type SrIrO_3 demonstrates the high conversion efficiency [1,2]. Of particular interest is the same perovskite-type CaIrO_3 (CIO), which has been known as a Dirac semimetal. Given the sensitivity of its transport properties to temperature and electric field, it is crucial to clarify spin current generation. In this talk, we present spin current generation in the Dirac semimetal CaIrO_3 .

The epitaxial CaIrO_3 film was grown on LSAT(001) substrate, fabricated by pulsed laser deposition. Figure 1(a) shows the X-ray diffraction pattern of $\text{CaIrO}_3(001)_{\text{pc}}$ film, where the pc represents pseudocubic. To investigate the transport properties, Hall coefficient and electrical resistivity of the 35 nm-thick-film were examined, as shown in Fig. 1(b). With increasing the temperature from 10 K to 300 K, the Hall coefficient exhibits a sign change, with positive below 100 K and negative above 100 K, while the electrical resistivity decreases. This behavior implies the semi-metallic feature of CIO. To evaluate the conversion efficiency, we conducted harmonic Hall measurements in Hall-bar devices consisting of ferromagnetic $\text{Co}_{20}\text{Fe}_{60}\text{B}_{20}$ and CIO bilayers. Figure 1(c) shows the angular dependence of the second-harmonic Hall resistance under in-plane external magnetic field at 0.2 and 1.5 T. The larger resistance with higher field suggests contribution of spin current generation stemming from CIO, providing the conversion efficiency of ~ 0.6 . Further investigation reveals a large reduction of the conversion efficiency with thinner thickness, indicating that the electronic structure characteristic of Dirac semimetallic CaIrO_3 is closely linked to spin current generation.



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Giant anomalous Hall effect from proximitized emergent field

Takahiro C. Fujita^{1,2}, Masashi Kawasaki^{1,2}

¹ *Dept. of Applied Physics, the University of Tokyo*

² *RIKEN CEMS*

Heterointerfaces based on transition metal oxides have been fascinating platforms for exploring a diverse range of exotic physical properties [1]. Among them, the discovery of a two-dimensional conducting interface formed at $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) has enabled the study of strongly correlated charge transport in two-dimensional electron systems (2DES) and the development of possible electronic device applications with novel functionalities. Another key finding at the LAO/STO interface is the observation of spin-dependent transport phenomena, such as spin-to-charge conversion and the anomalous Hall effect. Specifically, considerable effort has been devoted to attempting spin-polarized interfacial 2DES, for instance, by chemically doping magnetic elements into LAO or STO, inserting ferromagnetic layers between them, or replacing LAO with other magnetic perovskite oxides.

Recently, we have demonstrated that orthorhombic perovskite magnetic insulators are excellent sources of an emergent field (b_{em}) originating from their canted antiferromagnetic spin textures [2,3], wherein b_{em} denotes the Berry-phase-induced fictitious magnetic field experienced by conduction electrons. In particular, a huge effective magnetic field as large as ~ 30 T was observed in a $\text{CaRuO}_3/\text{DyFeO}_3$ (CRO/DFO) heterostructure [3], exhibiting an unconventional anomalous Hall effect (UAHE). This proximity effect of b_{em} provides a new design principle for perovskite oxide heterointerfaces.

In this work, we realize a spin-polarized 2DES in a DFO/LAO/STO heterostructure by leveraging the proximity effect of b_{em} (Fig. 1(a)). The DFO/LAO/STO heterostructure exhibits a stepwise UAHE resembling that of the DFO/CRO heterostructure but with a significantly larger Hall angle (θ_H) of $\sim 20\%$ at 2 K (Fig. 1(b)), which stems from the high carrier mobility in the 2DES ($\sim 2700 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$) [4]. This dramatic enhancement of the UAHE will pave the way to quantized Hall effect by b_{em} instead of applied magnetic fields, as theoretically proposed [5].

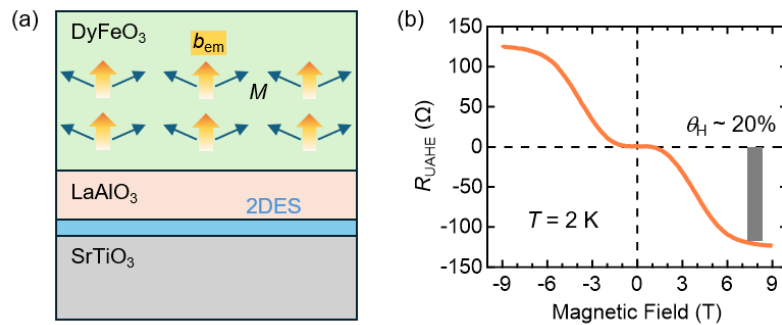


Figure 1: (a) Schematic of the DFO/LAO/STO heterostructure.

(b) Magnetic field dependence of the step-like unconventional anomalous Hall effect (UAHE) observed at 2 K.

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Electron Spin Resonance Simulation based on First Principles Calculation

Seongmun Kim¹, Yeongrok Jin², Jaekwang Lee^{1*}

¹*Department of Physics, Pusan National University, Republic of Korea*

²*The University of Tokyo, Japan*

Colloidal quantum dots (QDs) are promising light-emitting materials because they exhibit size-tunable emission, high color purity, and excellent solution processability. Among them, InP-based QDs have emerged as attractive alternatives to conventional Cd- and Pb-based QDs owing to their lower toxicity and potential for optoelectronic applications. In particular, the InP/ZnSe/ZnS core/shell structure has been widely studied because the ZnSe and ZnS shells improve luminescence efficiency and chemical stability. Nevertheless, these QDs still suffer from photodegradation under continuous illumination, which leads to a gradual decrease in emission efficiency and limits their practical use. Although defect formation has been considered one of the main origins of this instability, the microscopic pathway of defect generation and evolution has not been clearly identified.

In this work, we employ light-induced in situ electron spin resonance (ESR) spectroscopy together with density functional theory (DFT) calculations to investigate the formation and propagation of defects in InP/ZnSe/ZnS QDs under blue-light irradiation. Our results demonstrate that surface sulfur vacancies are first generated at the ZnS outer shell and subsequently act as initiation sites for defect evolution. These surface defects trigger the sequential formation of Zn-, S-, and Se-related vacancies, which propagate toward the ZnSe/ZnS interface and modify the electronic structure of the QDs. As a result, the band alignment changes from type-I to type-II, leading to reduced carrier confinement and a significant loss of external quantum efficiency. Notably, this defect evolution can be effectively suppressed by passivating surface sulfur vacancies, thereby improving photostability under prolonged illumination. These findings elucidate the microscopic origin of photodegradation in InP-based QDs and provide important insight into the design of highly stable and efficient quantum-dot optoelectronic devices.

References

Insight into the spin alignment in polycrystalline NiCo₂O₄ films

Shiho Sugiyama¹, Masaaki Tanaka^{1,2}, and Ryosho Nakane^{1,3}

¹ Department of Electrical Engineering and Information Systems

² Center for Spintronics Research Network (CSRN), ³ Systems Design Lab (d.lab),
The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, Japan

Ferrimagnetic NiCo₂O₄ (NCO) potentially having half-metallicity has emerged as a promising spin source for high-performance spin transport devices. In recent studies, high-quality epitaxial NCO films on a MgAl₂O₄ (MAO) substrate exhibit excellent material properties with almost perfect spin alignment via strong double-exchange interactions, where the suppression of defects has been revealed as a crucial issue [1, 2]. On the other hand, in polycrystalline NCO with many defects near grain boundaries, the local spin alignment almost certainly changes depending on spatially inhomogeneous defect density; however, its detailed characteristics and relationship with other characteristics, such as the saturation magnetization M_s , remain unclear. Here, we study the material properties of polycrystalline NCO films annealed at different temperatures from the aspect of multiple characteristics, mainly utilizing the resistivity-temperature (ρ - T) characteristics as a key probe that sensitively reflects the local spin alignment.

We prepared an NCO(36 nm)/MAO(1 nm)/Si(001) structure using our ultra-high vacuum electron-beam evaporator, and then annealed the samples in a 1-atm O₂ atmosphere at $T_{\text{ANL}} = 400$ and 500°C for 30 min. Figures 1(a)-(c) show the ρ - T characteristics measured under a constant applied current I (100 nA to 1 mA), where (a) corresponds to the as-deposited sample and (b) and (c) to the samples annealed at $T_{\text{ANL}} = 400$ and 500°C , respectively. For the annealed samples, the peak temperature T_p (arrow), which represents the local Curie temperature T_C' of defective regions [3][4], increases with increasing I . This indicates that increased spin injection from nearly defect-free grain bodies into defective near-grain-boundary regions enhances the degree of spin alignment there, increasing T_C' ($\sim T_p$), based on Ref. [3]. Regarding sample dependence, at 1 mA, T_p values are 164 K, 109 K, and below 4 K for the 400°C , 500°C , and as-deposited samples, respectively, indicating that the degree of spin alignment in near-grain-boundary regions after spin injection at 1 mA is higher in this order.

The sample dependence was analyzed in combination with M_s at 10 K and the nearest-neighbor hopping characteristic temperature T_{NNH} in the 250–290 K range at $I = 100$ nA. The underlying principle is that M_s reflects the volume fraction of grain-body regions and T_{NNH} reflects the initial spin alignment in near-grain-boundary regions at $I = 100$ nA. The 400°C sample has the largest fraction of grain body regions (smallest fraction of near-grain-boundary regions), since it has the largest M_s of $1.5\mu_B/\text{f.u.}$ compared with the as-deposited sample ($1.2\mu_B/\text{f.u.}$) and the 500°C sample ($0.7\mu_B/\text{f.u.}$). In addition, it can be concluded that the 400°C sample has moderate spin alignment near grain boundaries initially, since its T_{NNH} of 434 K is comparable to that (432 K) for the as-deposited sample and is smaller than that (567 K) of the 500°C sample. The integrated results consistently explain the fact that the 400°C sample has the highest degree of spin-injection-enhanced spin alignment near grain boundaries among the three samples when $I = 1$ mA.

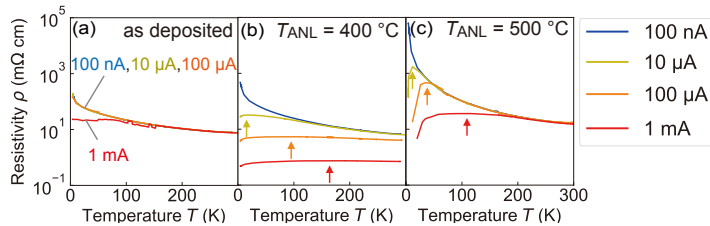


Figure 1(a)(b)(c): Resistivity-temperature characteristics (ρ - T) measured at various current I , where peaks are indicated by arrows: (a) as-deposited, (b) 400°C , and (c) 500°C . Note that 1-mA ρ - T curve has no clear peak within the measured temperature range 4–290 K.

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Anomalous Hall Effect and Interfacial Magnetic State in Compositionally-Graded Ru-substituted (La, Sr)MnO₃ Films

Yuto Abiko¹, Kenichi Kaminaga¹, Takayasu Hanashima², Kazuhiro Akutsu-Suyama², Shingo Maruyama^{1,3}, Hiroyuki Aoki^{4,5}, Yuji Matsumoto¹

¹ Department of Applied Chemistry, Tohoku Univ., Sendai, 980-8579, Japan

² Neutron Science and Technology Center, CROSS, Tokai, Ibaraki, 319-1106, Japan

³ Department of Electronics and Materials Science, Shizuoka Univ., Shizuoka, 432-8561, Japan

⁴ J-PARC Center, Japan Atomic Energy Agency, Tokai, Ibaraki, 319-1195, Japan

⁵ Institute of Materials Structure Science, KEK, Tokai, Ibaraki, 319-1106, Japan

The anomalous Hall effect (AHE), which is a transport phenomenon characteristic to ferromagnets, is sensitive to magnetization and spin-dependent scattering, and can thus be a useful electrical probe for spintronics. Among oxides which are ferromagnetic, (La_{2/3}Sr_{1/3})MnO₃ (LSMO) is a prototypical example for spintronic oxide materials owing to its robust ferromagnetism and high spin polarization. Recently, it was reported that the saturated anomalous Hall resistivity (ρ_{AHE}) increases with the concentration of Ru substituted for Mn in LSMO (LSMRO) [1]. On the other hand, we previously found that LSMRO films with linear Ru concentration gradients along the film-thickness direction exhibit ρ_{AHE} values different from those of uniformly doped films with the same average Ru concentration [2]. In this study, we investigate the relationship between this AHE response and the interfacial magnetic state through analyzing depth-resolved magnetization profiles of LSMRO films using polarized neutron reflectometry (PNR) [3].

100 nm-thick LSMRO films were prepared on LSAT(001) substrates by pulsed laser deposition [4]. They include two compositionally graded LSMRO films of UP (Ru 0→10%) and DOWN (Ru 10→0%), and uniformly doped LSMRO films with Ru concentrations of 0%, 5%, and 10% as reference samples. Two-channel PNR, which measures reflectivity for spin-up (R^+) and spin-down (R^-) neutrons to determine the depth-resolved magnetization profile, was performed at 100 K under an applied magnetic field of 1 T at BL17 SHARAKU, MLF, J-PARC.

According to Hall-effect measurements at 100 K, ρ_{AHE} was larger for the DOWN film than the UP film [2], reconfirming that the depth-dependent Ru concentration profile can contribute to the AHE response. The magnetic scattering length density (m-SLD) profiles obtained from reflectivity fitting in PNR analysis quantitatively revealed more or less reduction in magnetization of interfacial layers near the substrate interface as well as the film surface in the LSMRO films. Specifically, the magnetic moments near the substrate interface were estimated to be 1.50 $\mu_{\text{B}}/\text{f.u.}$ for the DOWN film and 1.16 $\mu_{\text{B}}/\text{f.u.}$ for the UP film, respectively. The value for the DOWN film was comparable to that of the uniformly doped Ru 5% film (1.56 $\mu_{\text{B}}/\text{f.u.}$). These results suggest that the AHE response is not determined solely by the average Ru concentration but is substantially correlated with the depth-dependent Ru concentration profile and the resultant magnetization distribution.

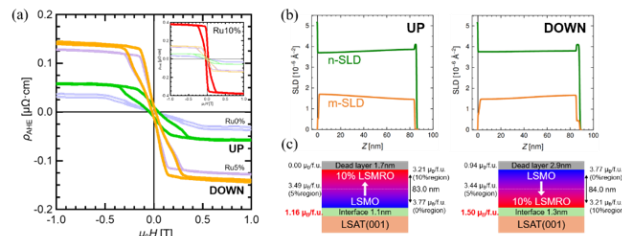


Figure 1: (a) Anomalous Hall resistivity vs. magnetic field for the LSMRO thin films. (b) Respective depth profiles of nuclear (nSLD, green) and magnetic (mSLD, orange) scattering length densities, extracted from PNR fits. (c) Schematic comparison of the layer-resolved magnetic structures derived from the PNR analysis.

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Self-driving sputter epitaxy of β -Ga₂O₃: from autonomous optimization to transferable oxide growth rules

Y. K. Wakabayashi^{1,*}, Y. Ogawa¹, F. B. Romero¹, T. Otsuka², and Y. Taniyasu¹

¹ Basic Research Laboratories, NTT, Inc., Atsugi, Kanagawa 243-0198, Japan

² Communication Science Laboratories, NTT, Inc., Soraku-gun, Kyoto 619-0237, Japan

*yuuki.wakabayashi@ntt.com

Self-driving laboratories (SDLs) accelerate materials development, yet most implementations remain black-box optimizers that yield limited transferable process knowledge. Here, we address this limitation in the context of β -Ga₂O₃ sputter epitaxy, where narrow growth windows have long hindered epitaxial growth. We developed a semiautonomous SDL platform for RF magnetron sputtering [Fig. 1a-1c]. Using Bayesian optimization (BO) with the Urbach energy (E_U) automatically extracted from optical transmittance as the optimization target, the system autonomously explored four growth parameters [substrate temperature, RF power, and Ar and O₂ gas flows] and identified high-quality heteroepitaxial growth conditions on 2-inch C-plane Al₂O₃, achieving $E_U = 182$ meV. Importantly, the BO-identified optimal heteroepitaxial growth conditions is directly transferable to β -Ga₂O₃ single-crystal substrates, yielding first the single-crystalline homoepitaxial growth of β -Ga₂O₃ by sputtering [Fig. 1d and 1e].

We further analyzed the BO-acquired dataset to extract human-usable knowledge for this thin-film growth. Using the dataset of 66 growth runs, we trained a random forest regression model that captures the E_U landscape on the four growth parameters. Our analysis showed that the substrate temperature is the dominant parameter, while the remaining parameters are largely additive; only the T -O₂ coupling defines the narrow window for high-quality growth. This structure suggests a human-usable optimization strategy: sequential one-dimensional tuning followed by final two-dimensional refinement in the T -O₂ plane.

Finally, we validated the extracted rules through human-guided re-optimization. Following the above optimization strategy, the process was further refined beyond the original BO result, reducing E_U from 182 to 163 meV. This result demonstrates that interpretable models can convert closed-loop data into actionable growth strategies that surpass the original black-box BO optimum. This work demonstrates that interpretable SDLs can advance oxide epitaxy beyond autonomous optimization by extracting transferable, human-usable growth knowledge.

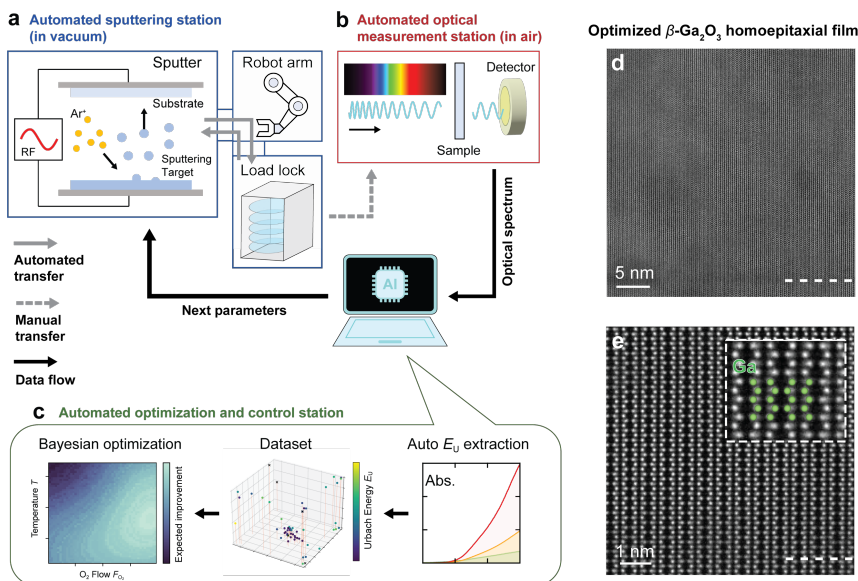


Figure 1: **a** Automated RF sputtering station. **b** Automated optical measurement station. **c** Automated optimization and control station that performs (1) auto-extraction of the E_U from the optical spectra, (2) dataset update, and (3) BO to propose the next growth conditions. **d** HAADF-STEM image of the homoepitaxial β -Ga₂O₃ film viewed along the [001] direction. **e** Magnified image near the interface (white dashed line) in **d**.

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Solid-Phase Epitaxial Growth of Rutile-type GeO₂ Thin Films Prepared by RF Magnetron Sputtering

Ali Shabab¹, Shohei Osawa¹, Daichi Oka¹, Yasushi Hirose¹

¹ Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University

Rutile-type GeO₂ (r-GeO₂) is a promising ultrawide-bandgap semiconductor ($E_g \sim 4.68$ eV) owing to its theoretically predicted ambipolar dopability and high carrier mobility [1]. However, the synthesis of single-crystalline r-GeO₂ thin films remains challenging due to energetic competition with amorphous and hexagonal GeO₂ phases, as well as the evaporation of GeO molecules at elevated temperatures. Recently, we demonstrated the solid-phase epitaxy (SPE) of r-GeO₂ [2], in which amorphous GeO₂ thin films deposited by pulsed laser deposition (PLD) at low temperature were crystallized by post-annealing to suppress evaporation of GeO. In this study, to further advance the SPE process toward scalable thin-film fabrication, we employed RF magnetron sputtering because of its superior compatibility with large-area deposition.

Amorphous GeO₂ thin films were deposited on TiO₂ (001) substrates at room temperature by RF magnetron sputtering using a GeO₂ target. A mixture of Ar and O₂ gases was introduced at a total pressure of 1.0 Pa. The partial O₂ pressure P_{O_2} was varied from 0.05 to 0.2 Pa to investigate the influence of oxygen stoichiometry on crystallization behavior. The amorphous thin films were subsequently annealed in air for 30 min using an infrared image furnace to promote crystallization.

X-ray diffraction (XRD) analysis confirmed that the amorphous GeO₂ thin films epitaxially crystallized into (001)-oriented r-GeO₂ upon annealing at ≥ 750 °C (Fig. 1). This crystallization temperature is comparable to that of amorphous GeO₂ films deposited by PLD [2], demonstrating the feasibility of applying the SPE process to sputter-deposited GeO₂ thin films. Although the influence of P_{O_2} was unclear from the XRD analysis, SEM observations revealed that low P_{O_2} caused spatial inhomogeneity in the crystallized films, probably due to the formation of the hexagonal phase (Fig. 2).

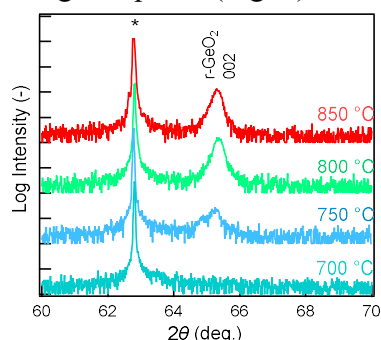


Fig. 1 2 θ - θ XRD patterns of the GeO₂ thin films annealed in air at various temperatures.

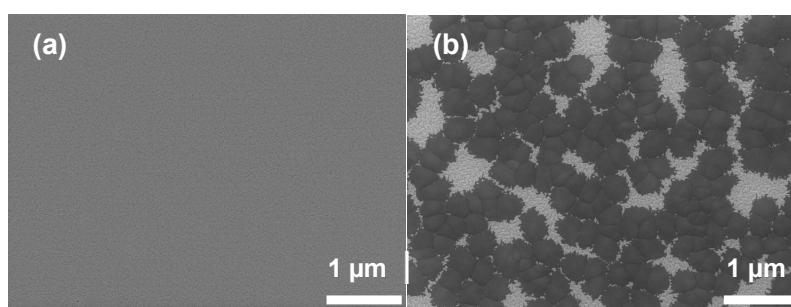


Fig. 2 SEM images of r-GeO₂ thin films crystallized at 800 °C from the amorphous films deposited under (a) $P_{O_2} = 0.2$ Pa and (b) 0.05 Pa. The bright and the dark grains in (b) are r-GeO₂ and impurity phase, respectively.

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Investigation on Ta doping in ultrawide bandgap semiconductor SrSnO₃

Haruka Shibata¹, Daichi Oka¹, Shunsuke Miyaji¹, Yasushi Hirose¹

¹ Tokyo Metropolitan Univ.

[Introduction] Perovskite-type SrSnO₃ is a semiconductor with an ultrawide bandgap of ~4.6 eV, which exhibits excellent transparent conducting properties upon electron doping [1]. While La doping at the Sr sites has been widely investigated, it faces a challenge where the carrier doping efficiency falls below 60% at doping concentrations of 1% or higher [2]. As an alternative donor impurity to La, Ta doping at the Sn sites is promising because the outermost 5d orbital of Ta is energetically far separated from the Sn 5s orbital and less hybridized with the conduction band minimum [3]. However, this approach has scarcely been investigated. In this study, we synthesized SrSnO₃ epitaxial thin films with 1% Ta doped at the Sn sites and investigated their physical properties.

[Experiment] The Ta:SrSnO₃ thin films were synthesized on SrTiO₃ (001) single crystal substrates by solid phase epitaxy. The precursor amorphous thin films were grown by pulsed laser deposition, where a SrSn_{0.99}Ta_{0.01}O_x target was irradiated by KrF excimer laser (wavelength: 248 nm, energy: 20 mJ, repetition rate: 10 Hz) in a vacuum chamber with an O₂ partial pressure of 5.0×10^{-2} Torr at room temperature. Subsequently, the films were crystallized by annealing in air at temperatures ranging from 900 °C to 1200 °C for 2 hours. Because the as-crystallized films were insulating, they were further annealed under a vacuum below 1.0×10^{-2} Pa at 790 °C for 30 minutes to induce electrical conductivity. The crystal structures were investigated by X-ray diffraction (XRD), and the electrical transport properties were measured with the van der Pauw technique at room temperature.

[Result and Discussion] In the XRD θ -2 θ patterns of the crystallized thin films, only the 00 l peaks of Ta:SrSnO₃ associated with the substrate peaks were observed for all the crystallization temperatures. In addition, epitaxial growth was confirmed by the spot peak of Ta:SrSnO₃ 103 asymmetric diffraction observed in reciprocal space mapping. Figure 1 shows the electrical conductivity as a function of the crystallization temperature for the Ta:SrSnO₃ thin films. The electrical conductivity of Ta:SrSnO₃ thin film crystallized at 1100 °C was significantly higher than that of the undoped SrSnO₃ thin film synthesized at the same condition. In addition, the Ta:SrSnO₃ thin film crystallized at 1000 °C, with the highest electrical conductivity, possessed a carrier density of 1.48×10^{20} cm⁻³, corresponding to the doping efficiency of 97.3%. These results indicated that Ta is an effective donor for SrSnO₃ as expected. However, the carrier mobility was limited to 2.97 cm² V⁻¹ s⁻¹ for the Ta:SrSnO₃ thin film crystallized under the optimum condition. This is probably attributed to the increased surface roughness caused by the high crystallization temperature. In the presentation, the effects of annealing conditions on the crystal structure, surface morphology, and electrical conduction properties will be discussed, along with another synthesis route using vapor phase epitaxy with a buffer layer to improve film quality.

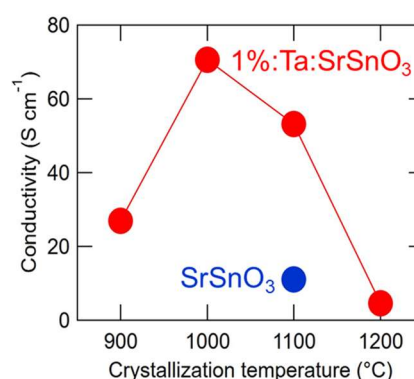


Figure 1. Crystallization temperature dependence of electrical conductivity for Ta:SrSnO₃ epitaxial thin films.

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Multinary Nitrides: A New Frontier for Oxide Epitaxy Techniques

Takuto Soma¹, Kodai Niitsu², Yushi Hiraide³, Mitsuki Matsuura¹, Kohei Yoshimatsu³, Akira Ohtomo³, Hiroshi Kumigashira¹

¹ IMRAM, Tohoku University, Sendai, Japan

² NIMS, Tsukuba, Japan

³ Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan

Over the past decades, sophisticated thin-film growth techniques in ultra-high vacuum—such as precision atomic-layer control, and combinatorial synthesis—have been extensively developed within the history of oxide epitaxy. These methodologies have driven the discovery of novel physics and functionalities in complex oxides. In this contribution, we propose that the legacy of oxide epitaxy techniques finds its most compelling new target in a non-oxide system: multinary nitrides.

Nitrides are an important class of materials; however, the high equilibrium vapor pressure of nitrogen poses a challenge for synthesizing bulk single crystals. Therefore, epitaxial thin-film growth is only the practical pathway to obtain the single crystals necessary for exploring their intrinsic properties and device applications [1]. Recent informatics approaches have predicted unexplored phases and functionalities in multinary nitrides [2], as evidenced by the experimental discovery of perovskite nitrides such as LaWN_3 and CeTa_3N_3 [3,4]. However, experimental material exploration remains sluggish, and the realization of their epitaxial films is exceedingly rare.

To break through this bottleneck, we have applied the combinatorial epitaxial synthesis technique—a powerful method pioneered and perfected in complex oxide research [5]—to multinary nitrides. To achieve high-crystalline films in a high-throughput process, we constructed a novel combinatorial sputtering system equipped with an ultra-high vacuum chamber ($\sim 10^{-7}$ Pa), a high-temperature substrate heater (~ 1000 °C), and program-controlled moving masks synchronized with multi-target reactive sputtering.

We first established precision of our system by growing high-quality binary Ta_xN_x epitaxial films on Al_2O_3 (0001) with high crystallinity and atomically abrupt interfaces (Fig. 1). Extending this to multinary systems, we successfully synthesized composition-spread La-Ce-Ta-N films (Fig. 2), mapping a vast phase space onto a single substrate. Through high-quality and high-throughput screening, we will showcase how the technical foundations built by the oxide-electronics community can powerfully accelerate the exploration of the uncharted multinary-nitride frontier.

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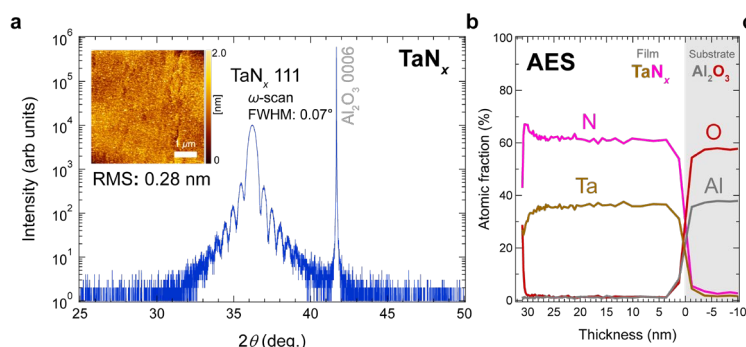


Figure 1 Epitaxial growth of high-quality Ta_xN_x thin films.

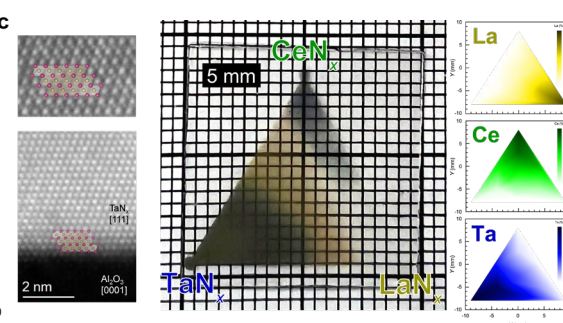


Figure 2 Combinatorial epitaxial growth of La-Ce-Ta-N thin films.

Solid Phase Epitaxy of Rutile-Type GeO₂ Films Under High Pressure

Keiji Shibata¹, Ryota Noda¹, Fumio Kawamura²,
Masashi Miyakawa², Hitoshi Yusa², Naoomi Yamada¹

¹ Chubu University, 1200 Matsumoto, Kasugai, Aichi 487-8501, Japan

²National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

Rutile-type GeO₂ (r-GeO₂) has been recently regarded as a promising next-generation power semiconductor. However, its crystal growth is challenging due to the close formation energies of the quartz and rutile phases, and its tendency to undergo thermal decomposition and sublimation at high temperatures and low pressures [1]. To overcome these issues, we developed a solid-phase crystallization method using high-pressure heat-treatment (HPHT) of amorphous GeO₂ (a-GeO₂) under GPa-scale pressures [2]. This method successfully yields single-phase r-GeO₂ thin films over a wide range of annealing temperatures (T_a) by stabilizing the rutile phase and suppressing thermal decomposition. In this study, we applied the HPHT method to the solid-phase epitaxy (SPE) of r-GeO₂ and obtained smooth r-GeO₂ SPE films on TiO₂(110) substrates.

A 300-nm-thick amorphous GeO₂ precursor film was reactively sputtered onto an atomically flat TiO₂(110) substrate from a Ge target without intentional heating. The sputtering was conducted in a mixed gas of Ar and O₂ (O₂/Ar = 1/9) under a total pressure of 0.5 Pa. The film was then encapsulated in a high-pressure cell and then subjected to HPHT using a belt-type high-pressure apparatus at a pressure of 4.5 GPa with temperatures ranging from 600 to 850 °C. For comparison, atmospheric fixed pressure heat treatment (APHT) was performed at 700 °C. X-ray diffraction was used for the confirmation of epitaxial growth.

HPHT at $T_a = 600\text{--}850$ °C yielded out-of-plane $\langle 110 \rangle$ -oriented r-GeO₂ films (Fig. 1). To verify the in-plane orientation, XRD ϕ -scan measurements of the 002 reflection from r-GeO₂ were conducted. As shown in Fig. 2, the ϕ scan exhibited two distinct peaks separated by 180°, reflecting the 2-fold rotational symmetry of r-GeO₂(110). This indicated single-domain SPE growth. Identical SPE crystallization occurred across the entire temperature range (600–850 °C), whereas no SPE was observed for the APHT sample.

Although success of SPE of GeO₂ in vacuum was recently reported, its temperature window is as narrow as $T_a = 700\text{--}750$ °C even with a buffer layer [3]. On the other hand, our high-pressure approach significantly expanded this temperature range. Which is attributed to the thermodynamic stabilization of the rutile phase under several GPa.

Acknowledgement

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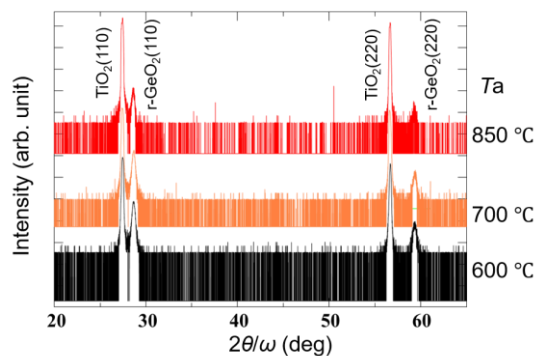


Fig. 1 Out-of-plane XRD patterns of GeO₂ films obtained through HPHT at 600 ~ 850 °C.

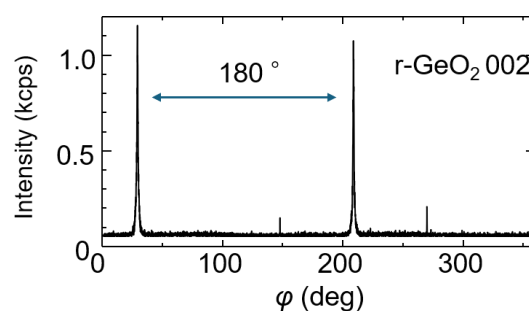


Fig. 2 In-plane ϕ -scan XRD pattern of r-GeO₂ film obtained through HPHT at 850 °C.

Epitaxial growth and electronic states of zirconium oxynitride thin films

Daichi Miyazaki¹, Kohei Yoshimatsu¹, Akira Ohtomo¹

¹ Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan

Metal oxynitrides provide additional degrees of freedom for oxide electronics through the introduction of shallow N $2p$ states and the increased flexibility of anion coordination [1,2]. Furthermore, these electronic states can be modulated in thin films through nonequilibrium growth and biaxial strain [2]. In this study, we focused on a homologous series of $\text{Zr}_{n+3}\text{O}_{2n}\text{N}_4$, which has remained unexplored in terms of epitaxial growth, despite its capability for bandgap engineering. We report on the successful epitaxial growth and electronic states of zirconium oxynitrides.

Zirconium oxynitride thin films were grown on yttria-stabilized zirconia (YSZ) (100) substrates using pulsed-laser deposition (PLD). A Zr_2ON_2 pellet synthesized via an ammonolysis process was used as the PLD target. The crystal structure, composition, and optical properties were investigated using X-ray diffraction (XRD), Auger electron spectroscopy (AES), and optical absorption spectroscopy, respectively. The projected density of states (DOS) was obtained using density functional theory calculations with QUANTUM ESPRESSO package.

Figure 1 (a) shows the out-of-plane XRD profile of the thin film grown at a substrate temperature of 400 °C under a nitrogen partial pressure of 1×10^{-4} Torr. The film reflection was observed at the low-angle side of the substrate reflection. The film exhibited the largest lattice spacing among the previously reported $\text{Zr}_{n+3}\text{O}_{2n}\text{N}_4$ phases. The full width at half maximum of the rocking-curve profile was less than 0.1° , indicating high crystalline quality [inset of Fig. 1(a)]. AES revealed a film composition close to $\text{Zr}_4\text{O}_5\text{N}_2$ ($n = 5$). Although an oxygen-rich composition was generally attributed to a larger bandgap (E_g), the film exhibited a smaller E_g of 1.95 eV [Fig. 1 (b)] than that of Zr_2ON_2 ($E_g = 2.6$ eV) [3]. A similar E_g trend was reproduced in the calculated DOS [Fig. 1(c)]. We found that the reduced E_g of $\text{Zr}_4\text{O}_5\text{N}_2$ originated from shallow unoccupied Zr $4d$ states associated with unique anion coordination environments in this phase. Our findings shed light on the hidden role of local anion coordination in the electronic states of oxynitrides.

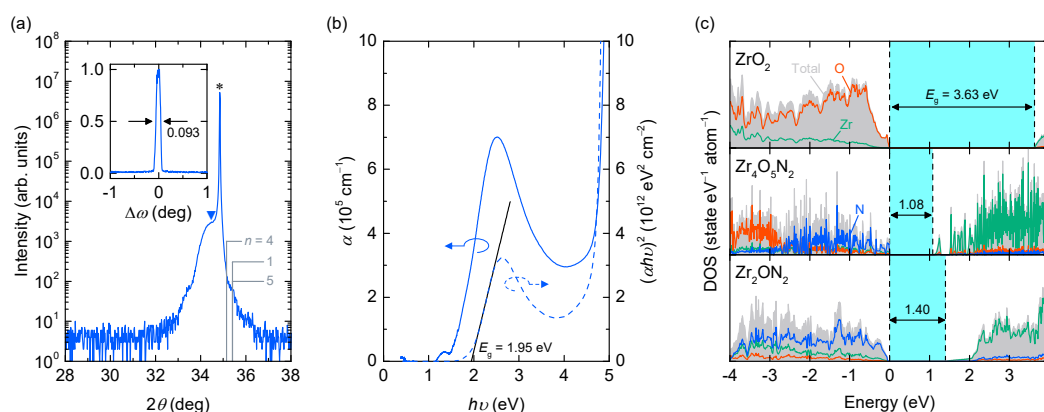


Figure 1: (a) Out-of-plane XRD profile of the zirconium oxynitride thin film. The asterisk (*) and triangle (▼) indicate reflections from the YSZ (100) substrate and thin film, respectively. The inset shows the rocking-curve profile of the thin-film reflection. (b) Optical absorption spectra (left axis) and a direct E_g estimated from the Tauc plot (right axis). (c) Projected DOS for ZrO_2 , $\text{Zr}_4\text{O}_5\text{N}_2$, and Zr_2ON_2 .

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Electronic Structure of Rocksalt YO Thin Films with Anomalous Valence States Studied by Photoelectron Spectroscopy

U. Aiba¹, D. Shiga^{1,2}, R. Hayasaka¹, S. Inoue¹, Y. Masutake¹, S. Hattori³, M. Negishi³, S. Sasaki³, T. Fukumura³, and H. Kumigashira^{1,2}

¹ IMRAM, Tohoku Univ., Sendai, Japan

² Photon Factory, IMSS, KEK, Tsukuba, Japan

³ Dept. of Chem., Tohoku Univ., Sendai, Japan

Since the transition metal Y has an electron configuration of $[\text{Kr}]4d^15s^2$, it typically forms the stable sesquioxide Y_2O_3 in the Y^{3+} state, and it is known that oxides with lower valence states such as Y^{2+} do not exist in nature. Recently, the successful synthesis of rocksalt-type YO thin films epitaxially grown on single-crystal substrates has been reported [1]. However, it remains unclear whether the Y ions in these films actually adopt the anomalously low valence state of Y^{2+} expected from their chemical composition [2]. In this study, we employed photoelectron spectroscopy to demonstrate the existence of the Y^{2+} state in single-phase YO thin films. Specifically, we identified the valence state through chemical shifts in the core-level spectra and directly observed the metallic state near the Fermi level (E_F), which is attributed to the Y $4d^1$ orbitals originating from this anomalously low valence state.

Figure 1(a) shows the Y $2p$ core-level spectra of YO alongside reference samples of Y-metal (Y^0) and sesquioxide Y_2O_3 (Y^{3+}). Clear chemical shifts were observed among the samples. A plot of the binding energies against the nominal valences [Fig. 1(b)] exhibits a clear linearity, identifying the Y valence in YO as $2+$. Furthermore, to verify the Y- $4d$ derived metallic state resulting from this Y^{2+} state, we measured valence-band spectra, as shown in Fig. 1(c). A clear spectral feature assigned to the Y $4d$ orbital is observed in the near- E_F region, and the presence of a distinct Fermi edge is confirmed. These results indicate that the Y ions in the YO thin films adopt the anomalously low valence state of Y^{2+} .

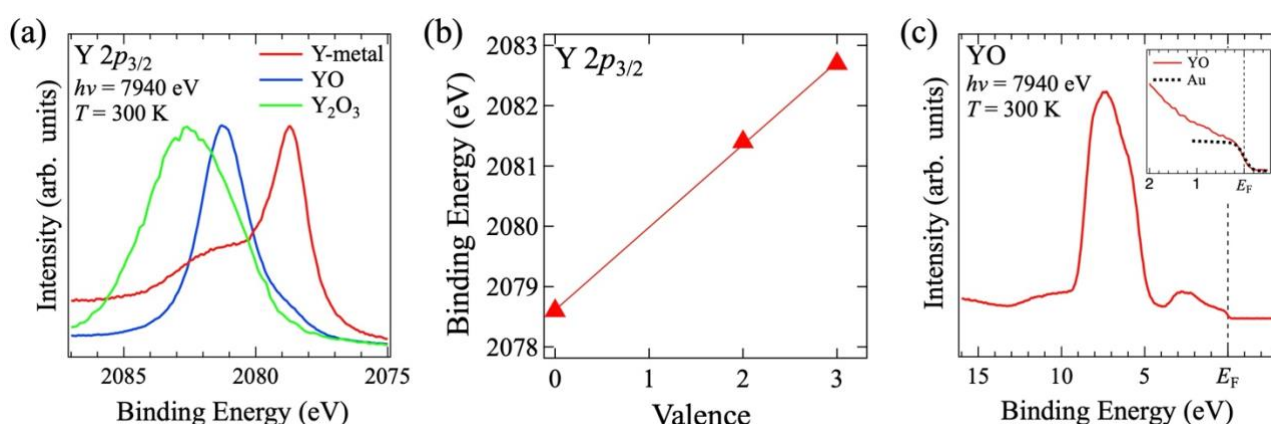


Figure 1: (a) Y $2p$ core-level spectra of Y-metal, YO, and Y_2O_3 . (b) Plot of the respective peak position as a function of nominal valence. (c) Valence-band spectra of YO. The inset shows an enlarged view of the region near E_F .

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Quaternary ultrawide-bandgap oxide semiconductor alloy: AlScGaO

Kazuki Koreishi¹, Kohei Yoshimatsu¹, Akira Ohtomo¹

¹ Department of Chemical Science and Engineering, Institute of Science Tokyo, Tokyo, Japan

Monoclinic ($C2/m$) $(\text{Al}_x\text{Ga}_{1-x})_2\text{O}_3$ (AlGaO), which is isostructural to $\beta\text{-Ga}_2\text{O}_3$, exhibits a tunable bandgap (E_g) ranging from 4.86 to 7.24 eV [1]. Silicon donors are predicted to be activated up to $x = 0.85$ in AlGaO ($E_g = 6.77$ eV) [1], making this ternary alloy a promising ultrawide-bandgap semiconductor for deep-/vacuum-UV transparent conducting oxides and high-voltage power electronics. However, achieving conductive AlGaO with high x is challenging since no conducting AlGaO with $x > 0.3$ has been reported to date. The possible origin of the lack of conductivity is strain relaxation in AlGaO epilayers grown on $\beta\text{-Ga}_2\text{O}_3$ substrates due to lattice mismatch [2]. To address this issue, we propose a quaternary alloy $(\text{Al}_x\text{Sc}_y\text{Ga}_{1-x-y})_2\text{O}_3$ (AlScGaO), because substitution of Ga with Sc can compensate for lattice mismatch between AlGaO and $\beta\text{-Ga}_2\text{O}_3$. Thus, co-substitution with an appropriate x/y ratio overcomes the limit of n -type doping imposed by lattice mismatch.

In this study, AlScGaO epilayers were grown on just-cut and 6° -miscut $\beta\text{-Ga}_2\text{O}_3$ (100) substrates using pulsed-laser deposition. We found that AlScGaO with composition $x/y = 1.5\text{--}3.2$ was nearly lattice-matched to $\beta\text{-Ga}_2\text{O}_3$ [3]. Electron energy-loss spectroscopy revealed that the E_g of 20-nm-thick epilayers with $x + y = 0\text{--}0.6$ could be tuned from 4.5 to 5.8 eV while keeping coherent heterostructures [Fig. 1(a)]. Furthermore, a coherently grown Si-doped AlScGaO ($x + y \sim 0.2$) thin film exhibited lower resistivity by 3 orders of magnitude than that of a strain-relaxed Si-doped AlGaO ($x \sim 0.3$) thin film [Fig. 1(b)]. These results demonstrate that the use of lattice-matched quaternary AlScGaO significantly improves transport properties, offering broad opportunities for exploring potential applications of ultrawide-bandgap oxide semiconductors.

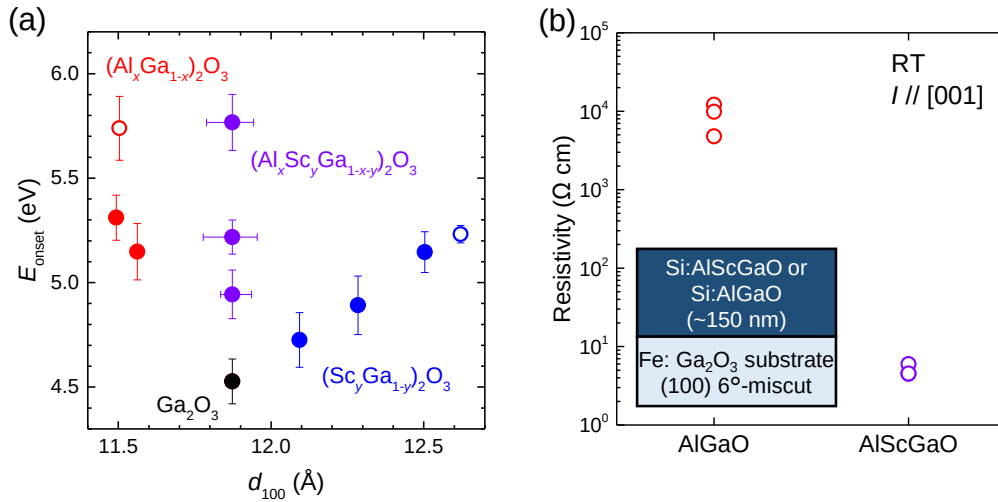


Figure 1: (a) E_g vs. out-of-plane lattice spacing d_{100} of AlScGO (purple). AlGaO (red) and ScGaO (blue) data are also plotted for comparison. The solid and open symbols represent coherently strained and partially relaxed epilayers, respectively. (b) Two-probe resistivity of strain-relaxed AlGaO ($x \sim 0.3$) and coherently grown AlScGaO ($x + y \sim 0.2$). Current was fed along the crystallographic [001] direction. The inset shows a schematic cross section of the epilayers for the resistivity measurements.

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Growth and Transfer of Hematite ($\alpha\text{-Fe}_2\text{O}_3$) Thin Films for Ultrafast Pump-Probe Spectroscopy

Fatma Ali

Fowlie lab(quoll-nu), Department of Materials Science and Engineering, Northwestern University

Ultrafast pump probe spectroscopy is a powerful technique for probing carrier dynamics in materials on femtosecond to picosecond timescales, but it requires high-quality, optically accessible thin films. Hematite ($\alpha\text{-Fe}_2\text{O}_3$) is a promising semiconductor for energy and optoelectronic applications due to its narrow bandgap (2.0–2.3 eV), chemical stability, non-toxicity, and earth abundance. However, transmission-based pump probe spectroscopy requires ultrathin, optically transparent freestanding hematite membranes compatible with our collaborator's experimental setup. This project addresses a key limitation in oxide thin film integration: extending water-soluble sacrificial layer transfer techniques, previously successful for perovskite oxides, to non-perovskite materials such as hematite.

In this work, hematite thin film membranes are grown using pulsed laser deposition (PLD) on LaAlO_3 substrates with water-soluble sacrificial layers, including $\text{Sr}_3\text{Al}_2\text{O}_6$ (SAO) and $\text{Ca}_3\text{Al}_2\text{O}_6$ (CAO)[1]. A thin SrTiO_3 (STO) layer is introduced to improve interfacial stability and provide a more favorable template for hematite growth, forming $\alpha\text{-Fe}_2\text{O}_3/\text{STO}/(\text{Sr,Ca})_3\text{Al}_2\text{O}_6/\text{LAO}$ heterostructures as shown in Figure 1. The membranes are subsequently released through dissolution of the sacrificial layer in water and transferred onto silicon nitride (SiN) substrates using a PMMA-assisted transfer process. Structural characterization using X-ray diffraction (XRD) and atomic force microscopy (AFM) is used to evaluate crystallinity, strain relaxation, domain size, and surface morphology.

A major challenge in this system is the large structural mismatch between hematite and the underlying perovskite layers. Hematite has a corundum structure, while LaAlO_3 is cubic, leading to strain, defect formation, and poor crystallinity, particularly on $\text{LaAlO}_3(001)$ [2]. To address this issue, this project focuses on $\text{LaAlO}_3(111)$, which provides improved pseudo-hexagonal symmetry matching with hematite and is expected to promote larger crystalline domains, lower surface roughness, and reduced defect density[2]. Preliminary work in the Fowlie Lab has already demonstrated successful growth of smooth STO/SAO/LAO heterostructure with low roughness, as well as successful membrane transfer processes. By optimizing substrate orientation, sacrificial layer composition, and PLD growth conditions, this work aims to establish reliable growth and transfer conditions for high-quality freestanding hematite membranes suitable for ultrafast optical characterization and future optical and quantum material applications.

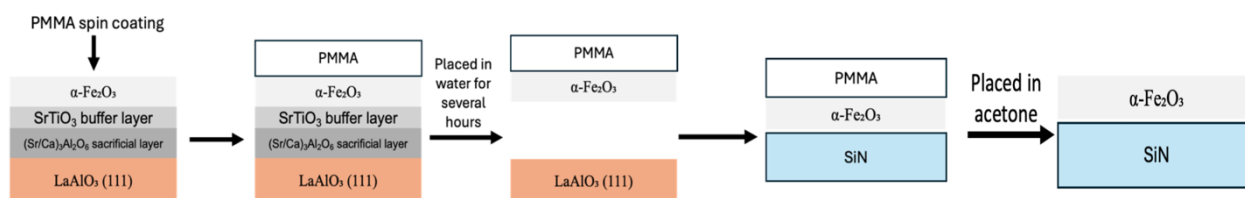


Figure 1: Schematic illustration of the membrane fabrication and transfer process.

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Sign reversal of the Hall coefficient in $\text{La}_2\text{DyNi}_2\text{O}_7$ thin films by varying epitaxial strain

Zhengyang Zhong¹, Motoki Osada¹, Atsushi Tsukazaki^{1,2}

¹ Department of Applied Physics and Quantum-Phase Electronics Center (QPEC),
University of Tokyo, 113-8656, Tokyo, Japan

² Center for Emergent Matter Science, RIKEN, Wako, Saitama, Japan

Superconducting signatures at 78 K were discovered in bulk $\text{La}_3\text{Ni}_2\text{O}_7$ under high pressure [1], which ignited tremendous research interest on Ruddlesden-Popper nickelates. Following that, ambient-pressure superconductivity with a transition temperature of 40 K has been achieved in compressively strained bilayer nickelate thin films [2,3]. This observation has been confined to the films on SrLaAlO_4 (SLAO) substrates, which impose a large compressive strain of approximately -2.0% . Subsequently, ambient-pressure superconductivity with a lower transition temperature of 10 K was observed in $(\text{La},\text{Pr})_3\text{Ni}_2\text{O}_7$ films grown on LaAlO_3 (LAO) substrates, which provide a smaller compressive strain of approximately -1.2% [4]. In the superconductivity in the bilayer nickelate films, epitaxial strain gives rise to significant effects involving deformations of the lattice and electronic structure.

In this study, we investigate the variations of lattice structure and resulting electronic structure by Dy substitution and applying various substrates. We fabricated $(\text{La},\text{Dy})\text{-Ni-O}$ films by pulsed-laser deposition on five types of substrates with different in-plane lattice constant: YAlO_3 (110), SrLaAlO_4 (001), LaAlO_3 (001), LaSrGaO_4 (LSGO) (001) and LSAT (001), to vary the in-plane epitaxial strain of films (Fig. 1(a)). While superconductivity appeared only in the film on SrLaAlO_4 , other films exhibited insulating behavior at below 10 K (Fig. 1(b)). The negative sign of Hall coefficient of the superconducting film was reversed in the films on tensile strain substrates such as LSGO (the inset of Fig. 1(b)), which provides insight into the electronic structure variation of bilayer nickelate thin films. In the presentation, detailed fabrication process and electrical transport properties will be discussed.

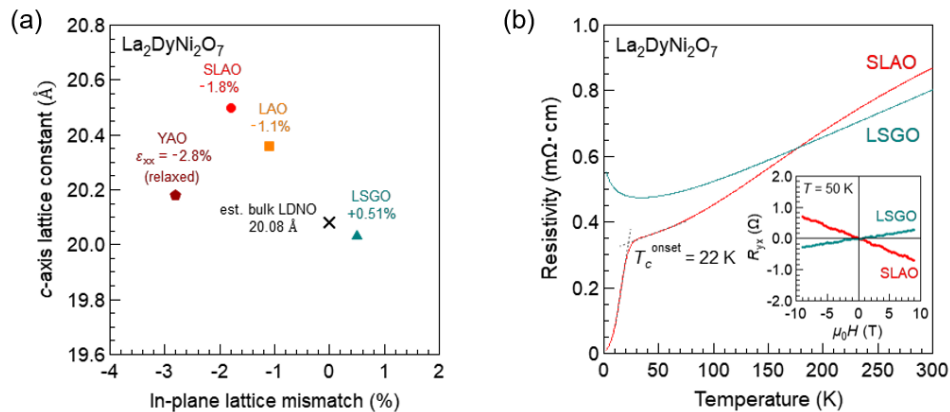


Figure 1: a, c-axis length as a function of in-plane lattice mismatch between the estimated bulk value of $\text{La}_2\text{DyNi}_2\text{O}_7$ and the results of the films. b, Temperature dependence of resistivity for the $\text{La}_2\text{DyNi}_2\text{O}_7$ films on SrLaAlO_4 (SLAO) and LaSrGaO_4 (LSGO). The inset shows magnetic field dependence of Hall resistance at $T = 50$ K for the films.

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Thickness-dependent low-temperature localization in CaRuO₃ ultrathin films

N. Obara¹, R. Yoshino¹, Y. Hamada¹, H. Nobukane², M. Sakoda¹

¹ *Dept. of Appl. Phys. Hokkaido Univ.*

² *Dept. of Phys. Hokkaido Univ.*

CaRuO₃ is a strongly correlated metallic oxide known to exhibit localized conduction in thin films [1] and thickness-periodic Mott insulating transitions in ultrathin films [2, 3]. Our previous experiments showed that an electric-double-layer transistor (EDLT) structure can induce a pronounced increase in resistivity in samples showing localization at low temperatures. This observation suggests that localized electronic states in CaRuO₃ ultrathin films are highly sensitive to electric-field-induced carrier accumulation. Thus, this study aims to clarify how low-temperature localization emerges in CaRuO₃ ultrathin films as a basis for understanding field-effect control of resistivity.

In this study, CaRuO₃ ultrathin films with different thicknesses were fabricated by molecular beam epitaxy (MBE), and their low-temperature transport properties were investigated. Figure 1 shows the temperature dependence of sheet conductance for films with different thicknesses. Even under optimized growth conditions based on previous work [4], thickness-dependent localization behavior was found to emerge at low temperatures. Further analysis of the localized samples revealed an approximately logarithmic temperature dependence of sheet conductance.

The logarithmic temperature dependence of conductivity is often observed in two-dimensional disordered conducting systems, where quantum corrections such as weak localization and electron-electron interactions become important. Furthermore, localization behavior appears to emerge when the sheet resistance is of the order of the quantum resistance h/e^2 , and this behavior strongly depends on the film thickness. This may be understood as a scenario in which the sheet resistance is enhanced near a Mott-insulating thickness in the thickness-periodic phase diagram, which may allow localization behavior to become observable. Based on analyses of magnetoresistance and temperature-dependent conductivity, we will discuss the origin of the logarithmic temperature dependence of conductivity by comparing possible contributions from weak localization, electron-electron interactions, and the Kondo effect.

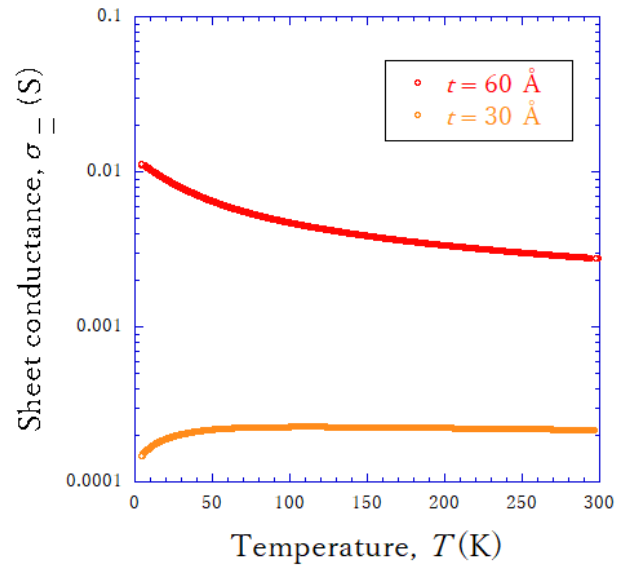


Figure 1: Temperature dependence of sheet conductance in CaRuO₃ ultrathin films

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Fabrication of Metastable Bixbyite Ce_2O_3 Thin Films

Mitsuki Yoshimura¹, Hiromichi Ohta²

¹ Graduate School of Information Science and Technology, Hokkaido Univ.

² Research Institute for Electronic Science, Hokkaido Univ.

Cerium oxide has attracted great attention in a wide range of fields, including catalysis and solid electrolytes. Homologous phases $\text{Ce}_n\text{O}_{2n-2}$ are formed depending on the oxygen deficiency concentration, and intermediate phases with $n \geq 7$, which are close to fluorite CeO_2 , have been extensively studied. In contrast, cubic bixbyite Ce_2O_3 with $n = 4$ is a metastable phase and thermodynamically unstable. Its fabrication is therefore challenging; it can only be obtained by heating a CeO_2/Ce bilayer interface under vacuum at high temperatures [1]. Our research group previously demonstrated that a $\text{Pt}/\text{SrCoO}_x/\text{CeO}_2/\text{YSZ}/\text{Pt}$ multilayer structure was effective for obtaining Ce_2O_3 phase by electrochemical reduction in air at 280°C [2, 3].

In this study, we successfully fabricated a Ce_2O_3 epitaxial thin film by removing the SrCoO_x layer with carbonated water (Figure 1). A bilayer film of $\text{SrCoO}_{2.5}/\text{CeO}_2$ was heteroepitaxially grown on a (001) YSZ single crystal substrate by PLD. Subsequently, Pt electrodes were deposited on the surface of the SrCoO_x thin film and the back surface of the YSZ substrate by DC sputtering. The fabricated $\text{Pt}/\text{SrCoO}_{2.5}/\text{CeO}_2/\text{YSZ}/\text{Pt}$ multilayer structure was heated in air at 280°C and a current was applied to electrochemically reduce the $\text{SrCoO}_{2.5}/\text{CeO}_2$. Finally, the Pt/SrCoO_2 layers were removed by immersion in carbonated water [4] to obtain a metastable Ce_2O_3 epitaxial thin film. The out-of-plane XRD pattern before removal of the Pt/SrCoO_2 layer confirmed a single-phase bixbyite Ce_2O_3 , with a lattice constant of 1.134 nm. Immediate XRD characterization after the removal of Pt/SrCoO_2 layers confirmed that partial oxidation had already occurred. Upon further exposure to air at room temperature, the film spontaneously oxidized back to a single-phase CeO_2 , with complete conversion occurring within 2.5 h. These findings provide an important foundation for elucidating the properties and functions of metastable Ce_2O_3 .

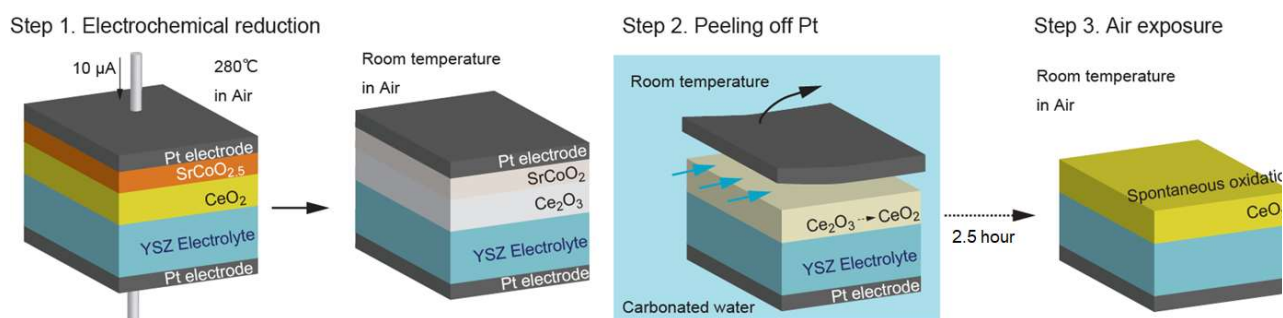


Figure 1: Schematic fabrication process of metastable bixbyite Ce_2O_3 epitaxial thin films.

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Atomistic Origin of Selenium-Enhanced Hole Transport in Amorphous Tellurium Oxides

Rafael Costa-Amaral¹ and Yu Kumagai^{1,2}

¹*Institute for Materials Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi, 980-8577, Japan*

²*Organization for Advanced Studies, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi, 980-8577, Japan*

High-mobility *p*-type amorphous oxides are difficult to realize because deep O-2*p* valence bands and structural disorder suppress hole transport. Oxygen-deficient amorphous tellurium oxides (*a*-TeO_x) recently challenged this view, with Se doping substantially improving thin-film transistor performance and hole mobility [1]. However, the atomistic role of Se remains unclear. Here, we use machine-learning-accelerated *ab initio* molecular dynamics and hybrid-functional electronic-structure analysis to clarify how Se modifies the conducting network in oxygen-deficient *a*-TeO_x. We first found that oxygen deficiency drives nanoscale segregation into Te-rich and oxide-like amorphous domains: the Te-rich network supplies valence-edge Te-5*p* transport states, while oxide-like/interfacial regions likely supply holes through Te vacancies [2].

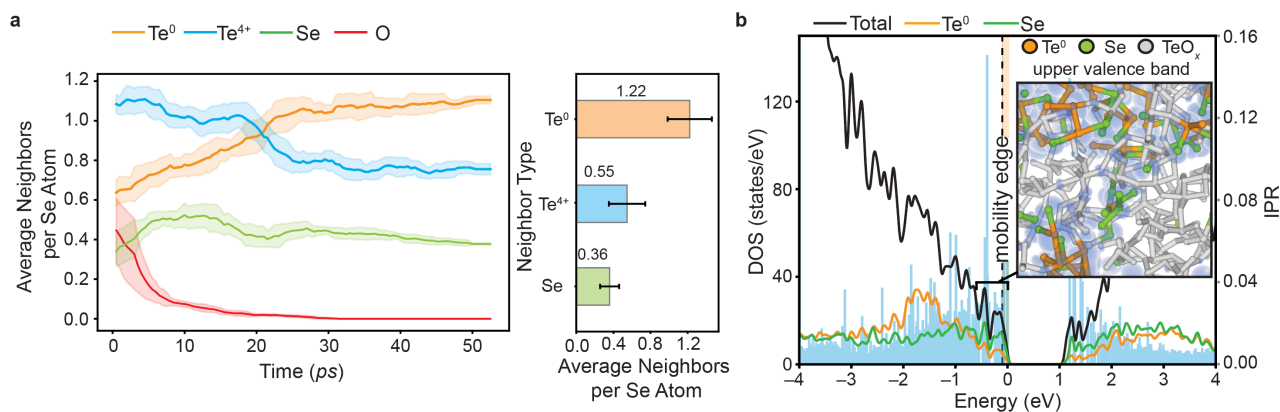


Figure 1: (a) Se-centered coordination during the melt-quench trajectory and after relaxation, showing preferential bonding to neutral Te rather than O. (b) Total/projected density of states (DOS) and inverse participation ratio (IPR) for Se-alloyed *a*-TeO_{1.2}, showing the Te-Se subnetwork contribution. The dashed line indicates the mobility-edge position, where electronic states become extended. Inset: isosurfaces of the upper valence band charge distribution.

For the experimentally optimized Se-doped composition (Se_{0.33}TeO₂), we show that Se does not incorporate randomly into the oxide matrix. Instead, Se preferentially alloys with the Te-rich domains: Se-O coordination rapidly disappears during the melt-quench trajectory, while Se-Te and Se-Se bonding persist. Coordination analysis (Fig. 1a) shows that Se is surrounded primarily by neutral Te neighbors (Te⁰), confirming its incorporation into the conducting subnetwork. Electronic-structure analysis further reveals that the upper valence band is dominated by the amorphous Te-Se network (Fig. 1b). Compared with undoped *a*-TeO_{1.2}, Se incorporation expands the valence-edge pathways and lowers the hole mobility-edge offset, from 0.75 to 0.20 eV on average, with some samples reaching values as low as 0.09 eV. These results indicate that Se enhances hole mobility not by acting as a conventional dopant, but by improving the connectivity and delocalization of the Te-based conducting network [2].

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Origin of the Narrow Low-Temperature Growth Window of PdCoO₂ in Molecular Beam Epitaxy

Mahsa Zamani¹, Gideok Kim¹, Michael Neumann¹, Bernhard Keimer¹

¹ *Max Planck Institute for Solid State Research*

Metallic delafossite PdCoO₂ has an interesting lattice structure, where a conducting layer of Pd¹⁺ is sandwiched between insulating [CoO₂]¹⁻ layers. This quasi-2D structure grants it highly anisotropic transport and a cryogenic mean free path surpassing 20 μm—the longest of any known metal [1]. Its remarkable in-plane conductivity has enabled sophisticated electrical transport studies such as quantum oscillations and directional probing of electrical currents. In addition, the high conductivity makes it an attractive candidate as a conducting element for electronics operating in oxygen-rich environments. However, growing high-quality thin films of PdCoO₂ has been a major challenge in the community.

There have been pioneering works using techniques such as PLD [2], sputtering [3,4], and MBE [5,6]. Among the three major PVD techniques used for PdCoO₂, the growth parameters used in MBE stand out: PLD and sputtering typically use temperatures of 700–800 °C, whereas MBE uses temperatures between 300 and 480 °C. Even with an uncertainty of up to 100 K in temperature due to difficulties in thermometry, this remains a significant gap.

Offering atomic-level control and an ultra-pure environment, MBE has been the premier deposition methodology. Oxidizing palladium under ultra-high vacuum has long been considered the main hurdle, which was overcome by using either ozone or atomic oxygen. However, despite these potent oxidants, single-phase synthesis remains restricted to an unusually narrow and low-temperature window [5,6]. Growth outside this regime leads to crystalline degradation or spontaneous decomposition into competing phases, indicating that subtle kinetic and thermodynamic constraints govern phase stability.

In this work, we uncover the fundamental mechanisms that constrain the growth window using MBE, in-situ RHEED, and X-ray diffraction. We demonstrate that the stability of the very first layer dictates the growth window, and we present a route to widen this window.

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Reactive sputtering growth of Sr₂IrO₄ thin films and Raman-based detection of iridium vacancies

Ksenia Rabinovich¹, Tim Priessnitz¹, Olfat Omareya¹, Maisam Abdallah¹, Kathrin Küster¹,
Bernhard Keimer¹, Gideok Kim¹,

¹ *Max Planck Institute for Solid State Research, Germany*

Among various transition-metal oxides, 5d compounds exhibit distinctive properties due to their sizable spin-orbit coupling (SOC) and extended *d*-shell orbitals. Sr₂IrO₄ (Sr-214) is an archetypal SOC-driven Mott insulator, where the spin ($S = 1/2$) and orbital ($L = 1$) angular momenta of the Ir⁴⁺ ions point in opposite directions and can be described in terms of a pseudospin $J_{\text{eff}} = 1/2$ [1]. As the electronic properties of Sr-214 are dominated by SOC, which links the lattice and spin degrees of freedom, the magnetic properties of Sr-214 are highly sensitive to lattice modifications such as strain [2].

Epitaxial strain is an obvious route to exploit this strain sensitivity. Nevertheless, growing epitaxial thin films of Sr-214 with high yield has remained challenging. The most widely used method for epitaxial growth is pulsed-laser deposition (PLD), in which a stoichiometric target is ablated by a focused excimer-laser beam. However, given the chemistry of iridium, PLD is not ideal because the high temperature generated during ablation can decompose the target into metallic iridium, hindering consistent growth.

We demonstrate the epitaxial growth of high-quality Sr-214 thin films using reactive off-axis sputtering. Conventional PLD of Sr-214 is often hindered by the volatility and decomposition of iridates, which promotes parasitic Ruddlesden–Popper phases and the formation of iridium vacancies. Owing to its fundamentally different vaporization mechanism, reactive sputtering overcomes these limitations and leads to reliable synthesis of iridate thin films.

By systematically tuning the growth parameters, we identified a well-defined window in which the Sr-214 phase is stabilized with minimal iridium deficiency. We further show that Raman spectroscopy can serve as a sensitive probe of point defects, consistent with our previous studies on various complex oxides [3–5]. Our analysis reveals that the concentration of iridium vacancies in Sr-214 increases progressively when films are grown outside the optimal window.

These results establish reactive sputtering as a robust and scalable route for integrating Sr-214 into oxide electronic and magnonic devices, and promote confocal Raman spectroscopy as an effective tool for characterizing point defects in thin films.

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Autonomous synthesis of metastable ferrites

M. Lippmaa¹, Y. Sun¹, H. Liang², I. Takeuchi², G. A. Kusne³

¹ *University of Tokyo, Institute for Solid State Physics, Chiba 277-8581, Japan*

² *University of Maryland, Dept. of Materials Sci. and Eng., College Park, MD 20742, USA*

³ *National Institute of Standards and Technology, Gaithersburg, MD 20899, USA*

We have developed an autonomous pulsed laser deposition (PLD) workflow for optimizing the growth process conditions of epitaxial oxide thin films [1]. In particular, we focus on metastable hexagonal $LnFeO_3$ (Ln : lanthanide) phases that can only be stabilized in thin film form due to epitaxial strain. Bulk materials with the same composition are known as orthoferrites and assume the orthorhombic structure.

The surface structure of the films was monitored by reflection high-energy electron diffraction (RHEED). The time-dependent RHEED image data was processed with a Cascade R-CNN convolutional neural network model for determining if a RHEED image represents a flat epitaxial layer, a three-dimensional transmission pattern, a polycrystalline or non-crystalline pattern, or a mixture of pattern types. The model reliably identified individual diffraction features, such as diffraction streaks or spots in complex diffraction patterns, allowing for periodicity analysis for the various diffraction pattern components. When multiple phases or crystal orientations coexist, multiple crystal lattice periodicities would be detected, allowing for near-real-time measurement of phase purity or composition. A quality metric was defined for characterizing each deposition run, combining phase purity, crystallinity, and phase stability information obtained from the RHEED images. A Gaussian process regression (GPR) model was used for iteratively selecting film growth conditions (temperature, oxygen pressure, pulse rate, and ablation energy) to optimize the quality metric. The autonomous synthesis workflow yields the optimal growth conditions and a multidimensional map of the process metric (film quality) over the whole parameter space.

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Orbital-Driven Lattice Softening and Anisotropic Spinodal decomposition in Rutile $\text{TiO}_2\text{-VO}_2$

Yuji Muraoka¹, Takayoshi Yokoya¹, Yoshito Takemoto²

¹ *Research Institute for Interdisciplinary Science, Okayama University, Japan*

² *Graduate School of Environmental, Life, Natural Science and Technology, Okayama University, Japan*

Spinodal decomposition is a phase separation process characterized by spontaneous and periodic concentration fluctuations. While this phenomenon is widely known in metallic alloys, spinodal decomposition in functional metal oxides can couple strongly with their underlying electronic structures. The rutile-type $\text{TiO}_2\text{-VO}_2$ metal oxide systems is well-known to exhibit unique decomposition characteristics: anisotropic decomposition along the c -axis, the formation of a nanometer-scale lamellar structure with alternating Ti-rich and V-rich layers (Figure 1), and decomposition occurring at a remarkably low temperature of 400°C [1,2], which corresponds to approximately one-third of their respective melting points.

Previously, this anisotropic decomposition was understood in terms of a macroscopic viewpoint, particularly regarding the minimization of elastic strain energy along the [001] direction [2]. However, the electronic background giving rise to this anisotropy and the microscopic origin enabling its operation at such low temperatures remain unaddressed.

This study focuses on the overlapping effect of V $3d$ orbitals (conduction bands) extending along the c -axis in rutile-type VO_2 and elucidate the anisotropic decomposition mechanism of this system. This orbital effect induces dynamic electronic screening along the c -axis, thereby softening the elastic constants [3]. We propose that the unique overlap of $3d$ orbitals inherent to VO_2 underlies the macroscopic anisotropy of the elastic strain energy. Even under TiO_2 solid solution conditions, this localized screening extends throughout the lattice as long as the VO_2 components maintain percolation networks. Furthermore, we hypothesize that the softening of elastic compliance due to this electron screening effect significantly lowers the elastic barrier (activation energy) associated with atomic motion and modulated structure formation along the c -axis, thereby enabling decomposition process even at 400 °C.

In the presentation, we will integrate the macroscopic perspective of elastic strain with the microscopic perspective of the electronic state to discuss the mechanism of selective phase separation along the c -axis in this system.

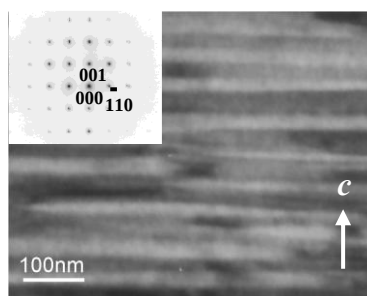


Figure 1: Electron micrographs of $\text{Ti}_{0.4}\text{V}_{0.6}\text{O}_2$ after annealing at 400°C for 24 h, taken along the [110] zone axis. Inset shows electron diffraction pattern of the electron micrograph of $\text{Ti}_{0.4}\text{V}_{0.6}\text{O}_2$.

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Anharmonic Phonon Effects from High-Order Force Constants

Kwanhong Park¹ and Jaekwang Leeⁿ (Times New Roman 12 pt)

¹ *1Department of Physics, Pusan National University, Pusan 46241, Republic of Korea*

Phonon dispersion relations characterize collective lattice vibrations in crystalline solids and provide fundamental insight into their vibrational properties. Within the harmonic approximation, which represents the quadratic expansion of the lattice potential energy around equilibrium, phonon dispersions can be obtained and are widely used to analyze basic lattice dynamical properties.

Nevertheless, the harmonic approximation does not incorporate phonon–phonon interactions and therefore cannot describe phonon scattering processes that are relevant to thermal transport phenomena such as lattice thermal conductivity. In addition, in temperature regimes where anharmonic effects become non-negligible, the resulting renormalization of phonon frequencies may require extensions beyond the harmonic framework.

In this work, we employ higher-order force constants to capture anharmonic phonon effects beyond the harmonic approximation. We investigate temperature-dependent phonon dispersions and analyze lattice thermal conductivity from first principles.

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Magnetotransport properties of EuTaO₃ epitaxial thin films

Hikaru Okuma¹, Yusuke Terada¹, Kazunori Ueno¹

¹ The University of Tokyo

Perovskite oxides with the formula ABO₃ possess a simple crystal structure and serve as a platform for researching electronic properties, as their physical parameters can be tuned by modifying the constituent elements at the A and B sites. In particular, the ferromagnetic and conductive perovskites EuMO₃ (M = Ti, Mo, Nb) [1-4] have been reported to exhibit a variety of interesting transport phenomena, including the anomalous Hall effect [1,2,4], the topological Hall effect [1], and a temperature(*T*)-dependent sign inversion of magnetoresistance (MR) [3,4]. On the other hand, in 5*d* transition metal oxides, the strong spin-orbit coupling (SOC) of 5*d* elements gives rise to numerous novel physical properties; for instance, SrTaO₃ exhibits one of the largest Rashba SOC among oxide materials [5]. In this study, as a new member of the 5*d* oxide family, we successfully fabricated conductive EuTaO₃ epitaxial thin films that possess both ferromagnetism and strong SOC. Furthermore, we found that by varying the underlying substrate and film thickness (*t*), the resistivity (ρ) of the thin films can be controlled within the range of 8×10^{-5} to $4 \times 10^{-3} \Omega \text{ cm}$, and that the MR transitions from negative to positive as ρ increases.

The EuTaO₃ epitaxial thin films were fabricated on insulating LSAT(100) and GdScO₃(110) substrates via PLD utilizing a KrF excimer laser. Laue fringes are clearly observed, indicating a flat interface (Fig. (a)). Figure (b) shows the ρ -*T* curves. On the LSAT substrate, ρ increases as *t* decreases. On the GSO substrate, which has a smaller lattice mismatch with the thin film than LSAT, ρ was reduced by approximately one order of magnitude. A distinct anomalous Hall effect was observed at 2 K (Fig. (c)), indicating the successful electrical detection of the ferromagnetic interaction between the localized Eu²⁺ 4*f* spins and the itinerant Ta 5*d* electrons. The MR changes from negative to positive with increasing R_s (Fig. (d)). This behavior is attributed to the localization of 5*d* electrons. Specifically, as the localization progresses, the weak antilocalization effect originating from the SOC becomes dominant over the suppression of magnetic scattering associated with the 4*f* and 5*d* ferromagnetic interaction. The EuTaO₃ thin films are relatively stable in ambient air, providing an excellent platform for exploring magnetism and transport phenomena driven by the interplay between SOC and ferromagnetism.

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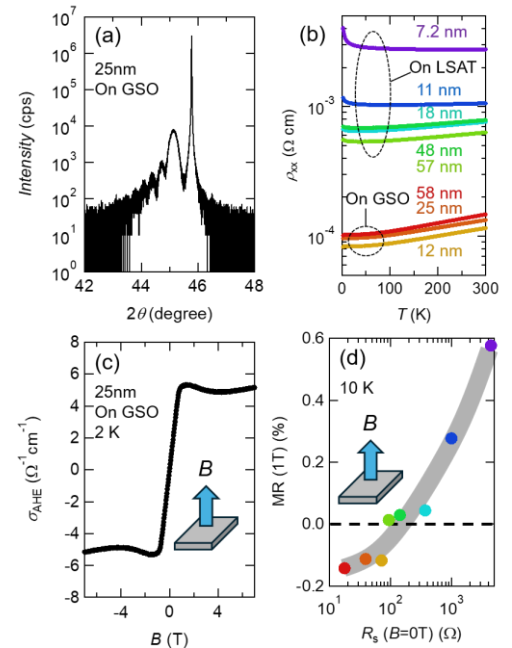


Fig. (a) Result of the out-of-plane $2\theta/\omega$ scanning with XRD for 25nm-thick EuTaO₃ film on GSO(110) (b) ρ -*T* curves for EuTaO₃ films (c) Anomalous Hall conductivity as a function of perpendicular magnetic field (d) Normalized MR versus R_s plots under perpendicular magnetic fields.

Electron doped CaTiO_3 films grown by gas source molecular beam epitaxy

Kurumi Hoshino, and Kei S. Takahashi

Department of Physics, Kanazawa University, Kanazawa 920-1192, Japan

Calcium titanate (CaTiO_3) is a perovskite-type oxide known as an incipient ferroelectric material such as quantum paraelectric. Quantum paraelectric perovskite oxides have attracted considerable attention, as electron doping induces a superconducting transition. This interest has been particularly heightened by the discovery that the quantum paraelectric SrTiO_3 exhibits superconductivity at around 0.3 K—with its transition temperature recently found to increase to 0.6 K under epitaxial strain [1]—and that the quantum paraelectric KTaO_3 undergoes a superconducting transition at 1–2 K upon interfacial electron doping at its (110) and (111) surfaces [2]. So far, there has been only a single report of metallic conduction in CaTiO_3 , achieved in bulk crystals [3]. Here, we identified electron doping of single-crystalline CaTiO_3 thin films as a challenging and intriguing research target.

In this study, CaTiO_3 thin films were epitaxially grown on (110)-oriented NdGaO_3 (NGO) substrates using gas-source molecular beam epitaxy (MBE). To investigate the optimal growth conditions, we systematically varied the substrate temperature and the titanium tetraisopropoxide (TTIP) flux. The structural properties of the films were characterized by X-ray diffraction (XRD) and atomic force microscopy (AFM).

In this presentation, we discuss the transport properties of optimally grown CaTiO_3 thin films electron-doped via oxygen vacancies and interfacial valence mismatch at the film/substrate interface.

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Influence of Growth Conditions on the Structural and Local Electronic Properties of Epitaxial Mn-Doped ZnO Thin Films

Shagun Thakur¹, Sulaiman Al Salem², Sreeju Sreekantan Nair Lalithambika², Jose de Jesus Velazquez-Garcia², Tobias Meyer¹, Toshiaki Ina⁵, Kiyofumi Nitta⁵, Iniyan Sivakumar¹, Jan Elsner^{3,4}, Christian Jooss¹, Simone Techert^{1,2}, Joerg Behler^{3,4}, Jasnamol Palakkal¹

¹ *Advanced Epitaxy, Institute of Materials Physics, Georg-August-University of Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany*

² *Deutsches Elektronen-Synchrotron, DESY, Notkestrasse 85, 22607, Hamburg, Germany*

³ *Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, 44780 Bochum, Germany.*

⁴ *Lehrstuhl für Theoretische Chemie II, Ruhr-Universität Bochum, 44780 Bochum, Germany.*

⁵ *Japan Synchrotron Radiation Research Institute/SPring-8, Hyogo 679-5198, Japan.*

Transition metal-doped ZnO thin films have attracted significant interest for optoelectronic, magnetic, and energy-related applications due to their tunable electronic structure and defect chemistry. This performance is strongly dependent on growth conditions, crystallographic structure, and dopant incorporation. Literature suggests that controlling oxygen vacancies and defects formation is crucial for tailoring properties relevant to optical, magnetic, and electrochemical processes [1,2].

In this work, epitaxial Mn-doped ZnO thin films were deposited by ion beam sputtering under either oxygen rich or oxygen deficient conditions. The films are predominantly oriented along the polar (0002) direction, with a minor contribution from the non-polar (11 $\bar{2}$ 0) orientation. Furthermore, additional in-plane rotational domains rotated by 30° are also observed. High-resolution transmission electron microscopy (HRTEM) confirms the dominant (0002) orientation with two in-plane rotational domains, indicating a strong influence of growth conditions on the resulting microstructure. To investigate the local atomic and electronic structure of the Mn doped ZnO films, X-ray absorption spectroscopy (XAS) measurements were performed. The combined HRTEM and XAS analysis demonstrates that the local electronic environment of Mn doped ZnO films are closely linked to the crystallographic orientation, which in turn is dependent on our growth conditions. [3] Preliminary OER measurements indicate that catalytic activity is dependent on crystallographic orientation and oxygen defect concentration. We observed that the oxygen-deficient sample was characterized by a dominant non-polar orientation with only a negligible polar contribution and exhibited enhanced OER activity. Conversely, the sample grown under oxygen-rich conditions showed both polar and non-polar orientations and showed poor electrocatalytic activity. We also observed significant degradation at higher applied voltages, which led to Zn dissolution from the films.

These results suggest that orientation-dependent effects may play a significant role in governing the functional properties of the material, especially for electrocatalytic activity. The observed correlation between crystallographic orientation and local electronic structure provides crucial insight into the design of transition metal doped Zinc oxide thin films for future optoelectronic, magnetic, and energy-conversion applications.

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WITHDRAWN

Crystal structure and fluoride-ion transport in Sr-substituted cerium fluoride thin films prepared by topochemical fluorination

Akira Chikamatsu¹, Ayuka Nakano¹, Miku Hagiwara¹, Dai Kutsuzawa², Erika Fukushi³, Hiroyuki Oguchi³, Fumihiko Uesugi⁴, Tsukasa Katayama⁵, and Yasushi Hirose⁶

¹ Department of Chemistry, Ochanomizu University

² Central Research Institute of Electric Power Industry

³ Chemistry and Materials Program, College of Engineering, Shibaura Institute of Technology

⁴ National Institute for Materials Science

⁵ Department of Applied Chemistry, Tokyo University of Science

⁶ Department of Chemistry, Tokyo Metropolitan University

Fluoride-ion batteries (FIBs) are attracting considerable attention owing to their high theoretical energy densities and the use of fluorine, which is far more abundant than lithium [1,2]. However, FIBs still suffer from poor charging-discharging characteristics and relatively high operating temperatures. Further fundamental research, including the development of new solid-state electrolyte materials, is therefore required to improve their performance. In this work, we investigated the crystal structures and fluoride-ion conductivities of Sr-substituted cerium fluoride epitaxial thin films—fluorite-type $\text{Sr}_{0.5}\text{Ce}_{0.5}\text{F}_{2.5}$ and tysonite-type $\text{Ce}_{0.975}\text{Sr}_{0.025}\text{F}_{3-\delta}$ —prepared by topochemical fluorination of epitaxial cerium oxide thin films, namely perovskite-type SrCeO_3 and fluorite-type $\text{Ce}_{0.975}\text{Sr}_{0.025}\text{O}_{2-\delta}$.

X-ray photoemission spectroscopy confirmed the reduction of Ce^{4+} to Ce^{3+} during fluorination, consistent with the formation of $\text{Sr}_{0.5}^{2+}\text{Ce}_{0.5}^{3+}\text{F}_{2.5}$ and $\text{Ce}_{0.975}^{2+}\text{Sr}_{0.025}^{3+}\text{F}_{3-\delta}$. Scanning transmission electron microscopy with energy-dispersive X-ray spectroscopy analysis of the $\text{Sr}_{0.5}\text{Ce}_{0.5}\text{F}_{2.5}$ film revealed complete transformation from the perovskite to the fluorite structure, accompanied by cation rearrangement in which Sr and Ce occupy the same crystallographic sites and fluorine replaces oxygen throughout the lattice. The resulting epitaxial films exhibited measurable fluoride-ion conductivity. Impedance spectroscopy of the $\text{Ce}_{0.975}\text{Sr}_{0.025}\text{F}_{3-\delta}$ films showed an in-plane conductivity of $1.1 \times 10^{-6} \text{ S cm}^{-1}$ at 300 K with an activation energy of 0.51 eV (Fig. 1). These results highlight the potential of cerium-based fluoride epitaxial films as model systems for elucidating F^- transport mechanisms.

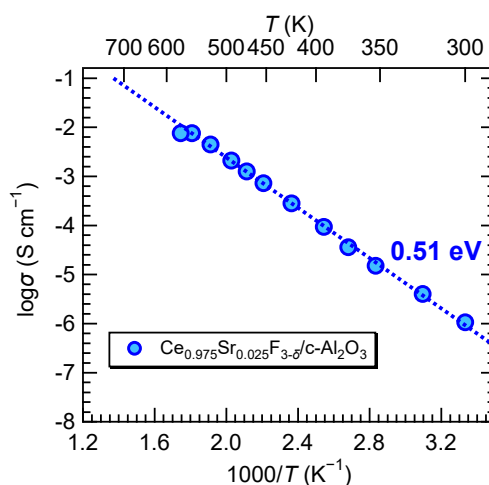


Figure 1: Arrhenius plot for the epitaxial $\text{Ce}_{0.975}\text{Sr}_{0.025}\text{F}_{3-\delta}$ on a c- Al_2O_3 substrate.

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Anion- and transition metal-dependent chemical trends in electronic structure parameters of 3d transition-metal compounds

Isao Ohkubo^{1,2}

¹ *Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS)*

² *Department of Materials Science, Institute of Pure and Applied Sciences, University of Tsukuba*

Advanced research on electronic structures of transition-metal oxides provides a foundation for understanding electronic structures of an entire class of ionic transition-metal compounds, as well as their unique physical and chemical properties. Irrespective of compound types, electronic structures and physical and chemical properties are typically controlled not only by changing transition-metal species but also by altering anion species and through partial substitution of these. To investigate anion species- and transition metal species-dependent on their electronic structures, we have calculated various electronic structure parameters (charge-transfer energy (Δ) and onsite Coulomb repulsion energy (U)) of over 500 binary and ternary ionic 3d transition-metal compounds possessing one of 11 types of anions (N^{3-} , O^{2-} , F^- , P^{3-} , S^{2-} , Cl^- , As^{3-} , Se^{2-} , Br^- , Te^{2-} , and I^-) [1]. Systematic chemical trends in variations of electronic structure parameters across anion species of 11 different families of 3d transition-metal compounds were comprehensively identified. Based on these analytical results, distinct relations between thermoelectric transport properties and electronic structure parameters were revealed. Furthermore, it was found that compounds with specific combinations of electronic structure parameters tend to exhibit high thermoelectric transport properties. A novel strategy was developed to identify promising candidate compounds with improved thermoelectric transport properties.

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Fabrication and Characterization of YBaFe₂O₅F Epitaxial Thin Films via Topochemical Fluorination

R. Tsuiji¹, A. Kamigaito¹, K. Shigematsu², H. Kumigashira³, T. Katayama⁴, Y. Hirose⁵,
A. Chikamatsu¹

¹Ochanomizu Univ., ²Science Tokyo, ³Tohoku Univ., ⁴Tokyo Univ. of Science,
⁵Tokyo Metropolitan Univ.

A-site double perovskite oxides $AA'B_2O_6$ (A, A' : alkali metal, alkaline earth metal, or rare-earth metal ions; B : transition metal ion) are transition metal oxides within the perovskite family that possess two crystallographically distinct metal ions at the *A*-site. Due to the layered ordering of *A*-site cations and periodically ordered oxygen positions, these compounds exhibit temperature-driven phase transitions accompanied by charge ordering and spin transitions. In particular, *A*-site double perovskite iron oxyfluorides ($AA'Fe_2O_5F$), in which oxygen vacancy sites are substituted by fluoride ions, have been predicted by first-principles calculations to exhibit coexisting ferroelectricity and magnetic ordering at room temperature [1]. However, to experimentally verify these predicted properties, the fabrication of single-crystal thin films of $AA'Fe_2O_5F$ is essential. Therefore, in this study, we have fabricated single-crystal YBaFe₂O₅F epitaxial thin films by pulsed laser deposition (PLD), followed by low-temperature topochemical fluorination, and have characterized the films crystal structure, electronic states, and dielectric properties.

YBaFe₂O_{5.5} precursor thin films were deposited on SrTiO₃ (001) (STO) substrates by PLD at a substrate temperature of 800 °C and an oxygen partial pressure of 1×10^{-6} Torr. The films were subsequently subjected to topochemical fluorination using polyvinylidene difluoride (PVDF) pellets at 200–350 °C for 12 h to obtain YBaFe₂O_xF_y single-crystal thin films. Film composition and O:F ratios were determined by energy-dispersive X-ray spectroscopy (EDS) peak-area ratios, while crystal structure was characterized by X-ray diffractometry (XRD) and reciprocal-space mapping (RSM). Fe valence states were probed by Fe 2p X-ray absorption spectroscopy (XAS), and dielectric properties of YBaFe₂O₅F film were measured using a precision LCR meter.

Figure 1 shows RSM images of the YBaFe₂O_{5.5} and YBaFe₂O₅F thin films. These results confirm that topochemical fluorination proceeds via extraction of oxide ions and insertion of fluoride ions while preserving the epitaxial strain. Figure 2 shows the frequency dependence of the relative permittivity and dielectric loss tangent ($\tan \delta$) of the YBaFe₂O₅F thin film at room temperature. Both quantities showed negligible frequency dependence over the range of 1–1000 kHz. The low $\tan \delta$ value of ~ 0.02 indicates that the film is a high-quality dielectric with minimal dielectric loss. The relative permittivity at 100 kHz was ~ 60 , comparable to that of the Fe³⁺-containing perovskite oxyfluoride SrFeO₂F [2].

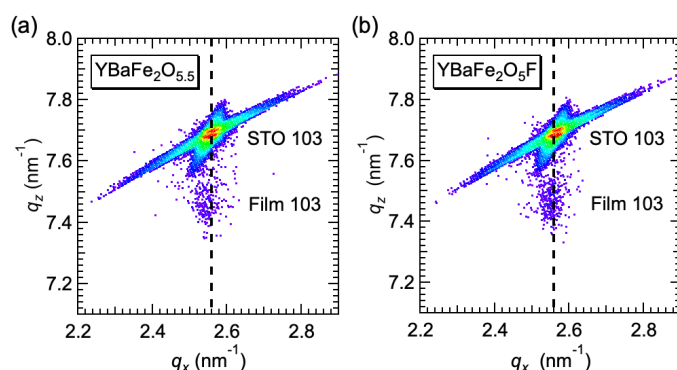


Figure 1. RSM images around the asymmetric 103 reflection for (a) the YBaFe₂O_{5.5} precursor film on STO substrates and (b) the YBaFe₂O₅F film.

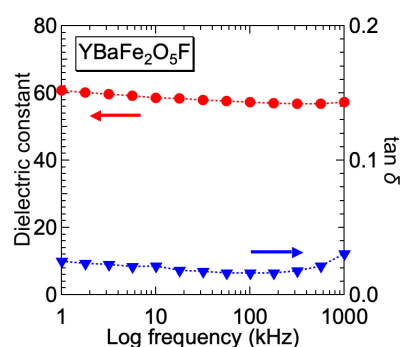


Figure 2. Dielectric constant and loss tangent of the YBaFe₂O₅F film.

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Hydrogenation-Induced Electrical Conductivity in LaAlO₃ Single Crystals

S. Toyooka¹, H. Okuma¹, T. Ozawa², K. Fukutani², Y. Nakamura¹, K. Ueno¹

¹ *Department of Basic Science, Univ. Tokyo*, ² *Institute of Industrial Science, Univ. Tokyo*

[Background] LaAlO₃ (LAO) is an insulating oxide with a rhombohedral perovskite structure at room temperature and a band gap of approximately 5.6 eV. Owing to its high dielectric constant and constituent material of oxide heterostructures, LAO has attracted considerable attention as a functional oxide material [1]. Despite this interest, inducing electron conduction in bulk LAO remains challenging. Although oxygen-vacancy formation and conventional impurity doping have been investigated in many oxides, their effectiveness of LAO is limited. Hydrogen is known to act as a donor and modify the electronic properties of oxides; however, hydrogen incorporation into LAO has been scarcely explored. Therefore, in this study, hydrogen plasma treatment, which provides a stronger reducing environment than conventional hydrogenation methods [2], was applied to LAO. As a result, successful hydrogen incorporation and indications of conductivity enhancement were obtained.

[Experimental Methods] LAO single-crystal substrates (Crystal Base Co., Ltd.) were hydrogenated by exposure to hydrogen plasma generated by applying a DC voltage in a hydrogen atmosphere. The samples were placed on the cathode and treated for 30 min. Structural characterization was performed using X-ray diffraction, elemental composition was analyzed by energy-dispersive X-ray spectroscopy, and the hydrogen depth profile was evaluated by nuclear reaction analysis (NRA).

[Results and Discussion] After hydrogen plasma treatment, the surface of the LAO substrate changed from transparent to gray. Figure 1 shows the hydrogen concentration depth profile obtained from hydrogenated LAO by NRA. Hydrogen incorporation was observed up to a depth of approximately 30 nm from the surface, with a maximum concentration corresponding to about 0.8 H atoms per unit cell. Figure 2 shows the time evolution of the electrical resistance of the hydrogenated LAO. Immediately after the hydrogen plasma treatment, the sample exhibited electrical conduction with a sheet resistance of approximately 100 k Ω /sq. However, the sheet resistance gradually increased upon exposure to air, and the sample reverted to an insulating state with a sheet resistance exceeding 20 M Ω within several days. These results suggest that hydrogen incorporated into interstitial sites acts as an electron donor, thereby inducing electrical conductivity in LAO. The increase in resistance is attributed to the out-diffusion and/or deactivation of hydrogen in air. The origin of the conductivity will be discussed on the basis of investigations of the stability of hydrogenated LAO under hydrogen atmospheres, transport-property measurements, compositional and surface-valence analyses, and theoretical calculations of the energetically favorable hydrogen configurations and the resulting electronic band structures.

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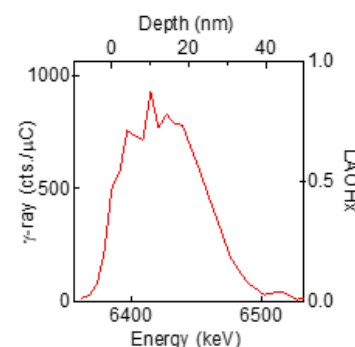


Fig1 Depth profile of hydrogen concentration in the sample measured by NRA.

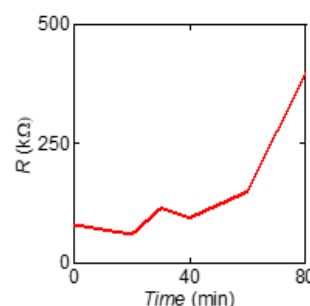


Fig2 Time dependence of the two-terminal resistance of the hydrogenated sample.

Capping-Layer Control of Topotactic Transformation in La–Sr Cobaltite Thin Films

Michal Kiaba¹, Jaeyeon Jo², Aliya Kaplan¹, Tareq Mufarech¹, Robert Klie², Jennifer Fowlie¹

¹ Materials Science and Engineering, Northwestern University, Evanston, Illinois, USA

² Physics Department, University of Illinois Chicago, Chicago, Illinois, USA

Ruddlesden-Popper (RP) cobaltites are promising layered transition-metal oxides with potential electronic, magnetic, and catalytic functionality. Because direct synthesis of phase-pure epitaxial RP thin films remains challenging, an alternative approach is topotactic transformation, converting a high-quality perovskite precursor into a lower-oxygen or layered phase through post-growth reduction. Previous work demonstrated that highly reducing anneals can transform epitaxial $\text{La}_{0.7}\text{Sr}_{0.3}\text{CoO}_{3-\delta}$ thin films into RP cobaltite phases through coupled oxygen loss and B-site cation redistribution.[1] Here, we build on this approach by introducing capping layers to control the topotactic transformation pathway and the associated cobalt redistribution, ultimately enabling the isolation of clean RP films.

We investigate the reduction-induced structural evolution of $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ epitaxial thin films using *in situ* and *ex situ* X-ray diffraction. Starting from the perovskite phase, oxygen loss drives a continuous transformation through an oxygen-vacancy-ordered Brownmillerite intermediate, ultimately resulting in the layered RP phase $\text{La}_{2-x}\text{Sr}_x\text{CoO}_4$. This transformation is accompanied by cobalt exsolution, producing cobalt nanoparticles at the film surface. While useful for supported nanoparticle catalysts, this exsolution is undesirable for fundamental studies of clean RP thin films.

To overcome this limitation, we employ different capping layers as a control mechanism for the topotactic process. Both insoluble (LaAlO_3) and water-soluble ($\text{Sr}_3\text{Al}_2\text{O}_6$) capping layers preserve the underlying structural phase transformation while confining the exsolved cobalt as it migrates to the film surface. Crucially, the $\text{Sr}_3\text{Al}_2\text{O}_6$ layer is easily dissolved in water post-reduction, cleanly carrying away the segregated surface cobalt. These results establish a robust route to epitaxial RP films and highlight the coupled roles of oxygen stoichiometry, cation mobility, and lattice topology in redox-driven oxide transformations.

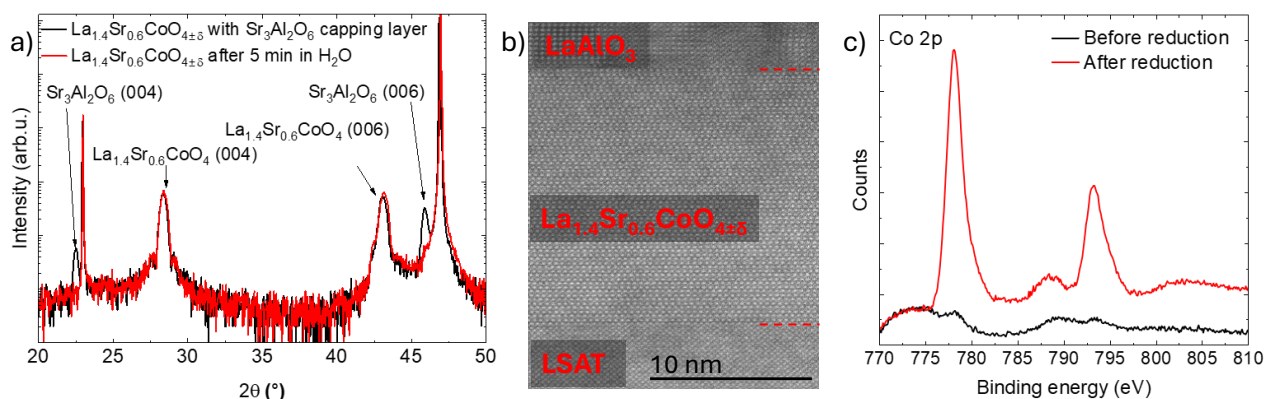


Figure 1. Structural and chemical characterization of transformed $\text{La}_{0.7}\text{Sr}_{0.3}\text{CoO}_3$ thin films. (a) XRD patterns of the RP film with a $\text{Sr}_3\text{Al}_2\text{O}_6$ capping layer before and after dissolution in water. (b) Cross-sectional TEM image showing sharp interfaces between LaAlO_3 , $\text{La}_{1.4}\text{Sr}_{0.6}\text{CoO}_4$, and LSAT after transformation. (c) Co 2p XPS spectra before and after reduction of sample with LaAlO_3 capping layer, showing cobalt exsolution during the transformation.

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Heterogenous Oxygen Sublattice Structure in $\text{La}_{0.67}\text{A}_{0.33}\text{MnO}_3$ (A=Ba, Ca, Sr) Thin Films

Roham J. Talei¹, Shihao Wei¹, Bruce A. Davidson², Aleksandr Yu. Petrov³, Nicolas Bonmassar¹

¹ *University of Stuttgart, Institute of Materials Science, Department for Materials Physics, Heisenbergstr. 3, 70569, Stuttgart, Germany*

² *Quantum Matter Institute, Univ. of British Columbia, 2355 East Mall, Vancouver BC V6T 1Z4, Canada*

³ *CNR-IOM, Area Science Park, Strada Statale 14, Km 163.5, I-34149, Trieste, Italy*

Despite identical nominal $\text{Mn}^{3+}/\text{Mn}^{4+}$ ratios at optimal doping $x=0.33$, the three canonical manganite compositions $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$, $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$, and $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ exhibit different Curie temperatures (T_C) of 340 K, 370 K, and 270 K, respectively.[1-3] This spread of 100 K remains quantitatively unexplained within mean-field double-exchange theory alone. The double-exchange transfer integral $t \propto \cos(\theta/2)$, where θ is the Mn–O–Mn bond angle, makes the oxygen sublattice geometry the decisive microscopic parameter: a bond angle approaching 180° maximizes d - p - d orbital overlap and therefore ferromagnetic bandwidth, while cooperative MnO_6 octahedral tilting and Jahn-Teller distortions bend this angle, locally suppressing double exchange. Conventional diffraction probes yield only spatially averaged bond angles and are inherently blind to the nanoscale heterogeneity of the oxygen sublattice arising from the different ionic radii of Ba^{2+} (1.36 Å), Sr^{2+} (1.18 Å), and Ca^{2+} (1.00 Å) relative to La^{3+} (1.03 Å). We propose that each of those dopants imposes a distinct local strain field and octahedral tilting pattern on the surrounding MnO_6 network.

Resolving this heterogeneity requires a real-space probe sensitive to oxygen positions at atomic resolution. We therefore grow epitaxial thin films of all three compositions by ozone-assisted molecular beam epitaxy (MBE), providing atomic-layer-precise control and a well-defined oxygen chemical potential during growth. Integrated differential phase contrast (iDPC) scanning transmission electron microscopy (STEM) imaging reveals a heterogeneous MnO_6 tilt distribution within each single composition. Critically, the degree of tilt heterogeneity scales inversely with T_C : broader bond-angle distributions reduce the percolation of ferromagnetic double-exchange pathways and systematically decrease the global ordering temperature.

By correlating these oxygen sublattice maps with electron energy-loss spectroscopic (EELS) maps of the local Mn oxidation state, we aim to unravel the direct microscopic link between A-site dopant identity, local oxygen geometry, and ferromagnetic exchange strength, identifying oxygen sublattice homogeneity as a previously unrecognised design principle for tuning T_C in doped manganites independently of carrier concentration.

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Microstructure and defect engineering driven tuning of the insulator–metal transition in RF sputtered VO₂ thin Films

K Gurukrishna¹, Amit K Rana², Shikhar Misra²

¹ *Department of Physics, School of Advanced Sciences (SAS), Vellore Institute of Technology (VIT) Vellore, 632014, Tamil Nadu, India*

² *Department of Materials Science and Engineering, Indian Institute of Technology Kanpur, Kanpur, Kalyanpur 208016, UP, India*

Vanadium dioxide (VO₂), a correlated oxide characterized by strongly coupled electronic and structural phase transitions, has emerged as a promising material for smart coatings and advanced electronic applications. In this work, we report the growth of VO₂ thin films by RF magnetron sputtering on Si (001), amorphous quartz, and c-cut α -Al₂O₃ (0006) substrates under varying sputtering powers. Through deliberate strain and microstructure engineering, we achieve broadband modulation of the metal–insulator transition (MIT), spanning more than 20 °C near room temperature. Nanocrystalline VO₂ films deposited on Si and quartz exhibit tailored polycrystalline microstructures, whereas epitaxial VO₂ layers on c-cut α -Al₂O₃ (0006) display pronounced out-of-plane compression. Our results demonstrate that epitaxial strain governs T_c tuning in VO₂ (020)/c-cut α -Al₂O₃ (0006) systems, while microstructural effects dominate transport in textured VO₂ films with preferred (011) orientation on Si and quartz. These findings are corroborated by X-ray diffraction, Raman spectroscopy, and electron microscopy. Importantly, precise control of strain and microstructure through sputtering power provides a scalable pathway toward VO₂-based functional devices.

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Stabilizing oxygen stoichiometry in BaFeO₃ thin films by capping layer and cold-temperature storage

Aliya Kaplan¹, Antoine Devinenti¹, Michal Kiaba¹, Jennifer Fowlie¹

¹ *Materials Science and Engineering, Northwestern University, Evanston, Illinois, United States*

Helimagnetic materials are hosts to phenomena such as skyrmions and magnetoelectricity, which are of significant interest both for exploring fundamental questions in topological magnetism as well as enabling advances in low-power spintronics, high-density data storage, and quantum sensors. Helimagnetism typically occurs as a result of magnetic frustration or the Dzyaloshinskii-Moriya interaction, which requires broken inversion symmetry. As a centrosymmetric cubic perovskite, the reported helimagnetism in BaFeO₃ powders is better attributed to a modified double exchange mechanism arising from negative charge transfer energy [1]. Investigating helimagnetism in the alkaline-earth ferrites is complicated by the tendency of Fe⁴⁺ to reduce to the more stable Fe³⁺ state, driving oxygen loss as a charge-compensation mechanism. Because oxygen vacancies alter exchange interactions, even small stoichiometric deviations (BaFeO_{3-x}) are expected to affect magnetic order. In thin films, this challenge is magnified due to the high surface-to-volume ratio. Utilizing post-growth annealing in ozone, capping layers, and storage on dry ice to mitigate oxygen loss, we have produced the highest quality and most stable BaFeO₃ films to date. This advancement in the synthesis and stability of BaFeO₃ thin films enables exploration of previously inaccessible fundamental questions regarding the magnetic properties of this system.

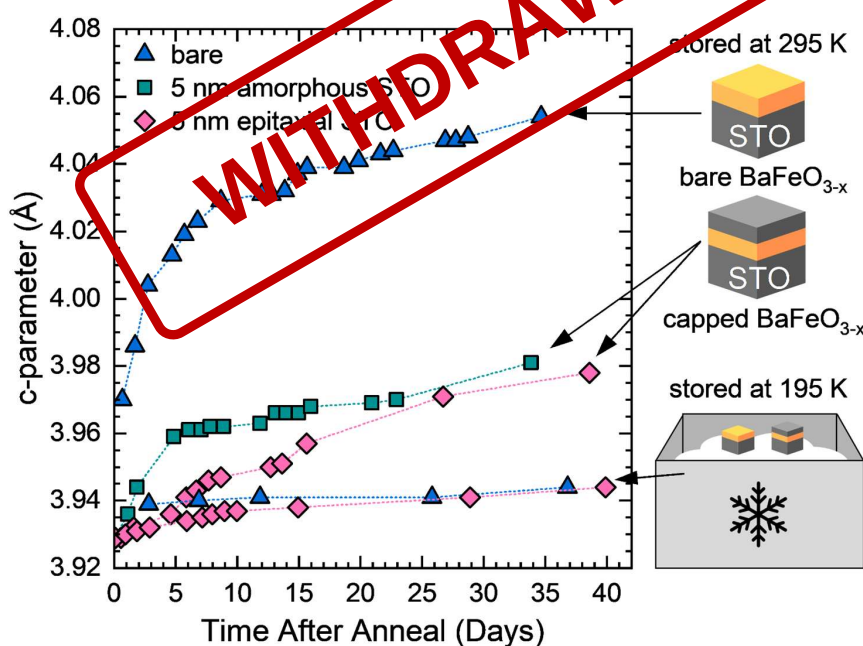


Figure 1: a) Time-dependence of c-parameter over several weeks. Bare BaFeO_{3-x} shows significant c-parameter expansion over time, indicating the formation of oxygen vacancies. BaFeO₃ films, both bare and capped with epitaxial SrTiO₃, when stored on dry ice, become stable over several weeks.

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Thermal conductivity of In_2O_3 thin films

^oHikaru Sadahira¹, Mitsuki Yoshimura¹, Prashant Ghediya², Yusaku Magari², Hiromichi Ohta²

¹ Graduate School of Information Science and Technology, Hokkaido University, Sapporo, Japan

² Research Institute for Electronic Science, Hokkaido University, Sapporo, Japan

Indium oxide (In_2O_3) has been intensively studied as the active layer of thin-film transistors^[1-3], because its field-effect mobility ($\mu_{\text{FE}} > 70 \text{ cm}^2/\text{Vs}$) is higher than that of the widely used oxide semiconductor InGaZnO_4 ($\sim 10 \text{ cm}^2/\text{Vs}$) and meets the mobility required for driving devices of next-generation 8K displays. However, the bulk thermal conductivity of In_2O_3 ($\kappa \sim 15 \text{ W/mK}$ ^[4]) is one order of magnitude lower than that of low-temperature poly-Si ($\kappa \sim 150 \text{ W/mK}$), which may pose a problem for heat dissipation in practical applications. In this study, we measured the thermal conductivity of polycrystalline In_2O_3 films on alkali-free glass substrates and epitaxial In_2O_3 films on (111) YSZ substrates. The thermal conductivity of the polycrystalline films was found to be about half that of the epitaxial films. All films were prepared by pulsed laser deposition (PLD)^[2,3,5], and the thermal conductivity was measured at room temperature by time-domain thermoreflectance (TDTR). X-ray diffraction (resolution: 20 nm) revealed that the crystallite size of the films on glass was comparable to the film thickness and increased with film thickness (Fig. a). In contrast, spiral grains with a step-and-terrace structure were observed on the surface of the films on (111) YSZ substrates. Figure b shows the film-thickness dependence of the thermal conductivity of the In_2O_3 films. Assuming an interfacial thermal resistance of $1 \times 10^{-9} \text{ m}^2\text{K/W}$ and extrapolating based on Matthiessen's rule, the bulk thermal conductivity was determined to be $\sim 4 \text{ W/mK}$ for polycrystalline In_2O_3 and 9 W/mK for epitaxial In_2O_3 on (111) YSZ.

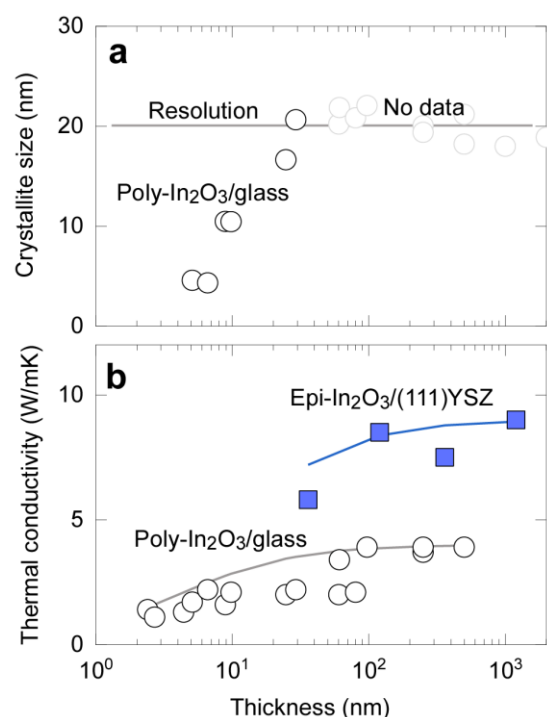


Fig. a | Thickness dependence of crystallite size for the polycrystalline In_2O_3 thin films measured by XRD (resolution limit: $\sim 20 \text{ nm}$).

Fig. b | Thickness dependence of thermal conductivity for the polycrystalline In_2O_3 thin films and epitaxial In_2O_3 thin films. The solid lines indicate Matthiessen's rule. The boundary resistance of $1 \times 10^{-9} \text{ m}^2\text{K/W}$ was used.

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Thermal-Stress Design in Quasi-2D Semiconductors via Anisotropic Thermal Expansion Engineering

Y.H. Lee¹, Y.J. Lin², P.W. Lin², W.T. Chen³, C.M. Hsu⁴, P.L. Liu⁴, Hiromichi Ohta⁵, Y.H. Chu¹.

¹Department of Materials Science and Engineering, National Tsing Hua University

²Department of Materials Science and Engineering, National Yang Ming Chiao Tung University

³College of Semiconductor Research, National Tsing Hua University

⁴Graduate Institution of Precision Engineering, National Chung Hsing University

⁵Research Institute for Electronic Science, Hokkaido University

Quasi-2D semiconductors are promising for next-generation electronics due to their layered structures, tunable properties, and substrate compatibility. However, thermal cycling induces stress from thermal expansion coefficient (CTE) mismatch with substrates. Here, we show that substrate choice and composition critically determine the thermal stress state in quasi-2D thin films. We systematically quantify the in-plane and out-of-plane CTE of the Bi₂O₂X (BOX, X = Se, S, Te) family, providing guidelines to minimize thermal stress.

We first establish intrinsic thermal expansion using freestanding films and first-principles calculations. Density functional theory predicts in-plane CTE on the order of 10⁻⁶ K⁻¹ and out-of-plane CTE on the order of 10⁻⁵ K⁻¹. The C/A ratio remains nearly constant (~3.1), reflecting intrinsic anisotropy. Freestanding measurements confirm stable C/A ratios (~2.9) across all compositions, serving as a reference free from substrate constraint.

Using this intrinsic benchmark, we analyze BOX films on SrTiO₃ (STO) and Si substrates. On STO, the C/A ratio varies significantly (2.6–3.8), indicating unstable anisotropic expansion and strong substrate-induced stress. In contrast, BOX films on Si exhibit nearly constant C/A ratios (~3.4), suggesting a more uniform anisotropic response and reduced thermal stress.

These results reveal that substrate choice plays a decisive role in governing thermal expansion in quasi-2D films: STO induces stronger thermal stress, while Si enables more stable anisotropic expansion. By using deviations from intrinsic anisotropy as a quantitative metric, we establish a unified framework for diagnosing thermal stress in quasi-2D semiconductors, providing practical guidance for substrate selection and device integration.

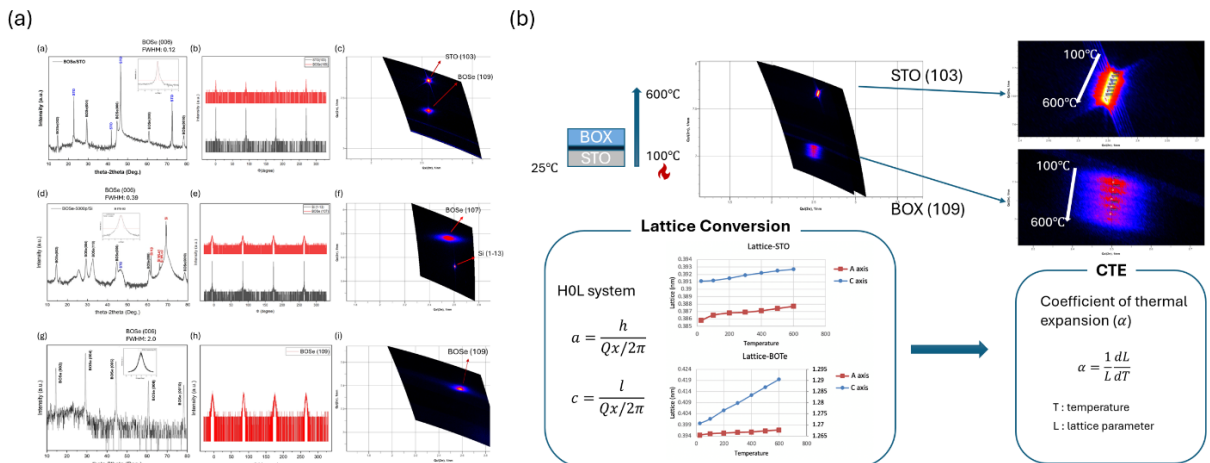


Figure 1: (a) X-ray structural characterization of BOX thin films on STO, Si, and freestanding configurations. (b) Determination of CTE by in-situ high-temperature reciprocal space mapping

Effect of Annealing Atmosphere on Structure and Electrical Properties of β -Ga₂O₃ Thin Films

Suyue Chen¹, Yixuan Li¹, Weitao Fan², Wei Ren², Weili Wang³

¹ School of Electronic Engineering, Xi'an University of Posts & Telecommunications, Xi'an China

² School of Science, Xi'an University of Posts & Telecommunications, Xi'an China

³ School of Physical Science and Technology, Northwestern Polytechnical University, Xi'an China

Monoclinic β -Ga₂O₃ is an ultra-wide bandgap semiconductor (~ 4.8 eV) with high breakdown field, promising for deep-UV detectors and high-power devices[1,2]. Annealing-induced Si diffusion from quartz substrates enables effective n-type doping[3,4], yet the role of atmosphere remains unclear. Here, β -Ga₂O₃ films were deposited on quartz by electron beam evaporation and annealed at 500 °C in Ar, air, and O₂. Structural, morphological, chemical, electrical, and optical properties were systematically investigated. XRD confirms polycrystalline β -Ga₂O₃; O₂-annealed films show the largest grains (21.88 nm). SEM reveals cracked surfaces in Ar and dense morphology in O₂. XPS indicates air annealing yields the highest Ga³⁺ fraction (73.11%) and moderate oxygen defects. Hall measurements show the air-annealed sample achieves a carrier concentration of $6.56 \times 10^{18} \text{ cm}^{-3}$ and resistivity of $0.0297 \Omega \cdot \text{cm}$, two orders of magnitude higher than Ar- and O₂-annealed counterparts ($\sim 10^{16} \text{ cm}^{-3}$). Moderate oxygen partial pressure in air promotes active SiO diffusion from quartz and suppresses Si-O_x cluster formation, enabling efficient substitutional doping. This work clarifies atmosphere-regulated Si doping and provides guidance for high-performance β -Ga₂O₃.

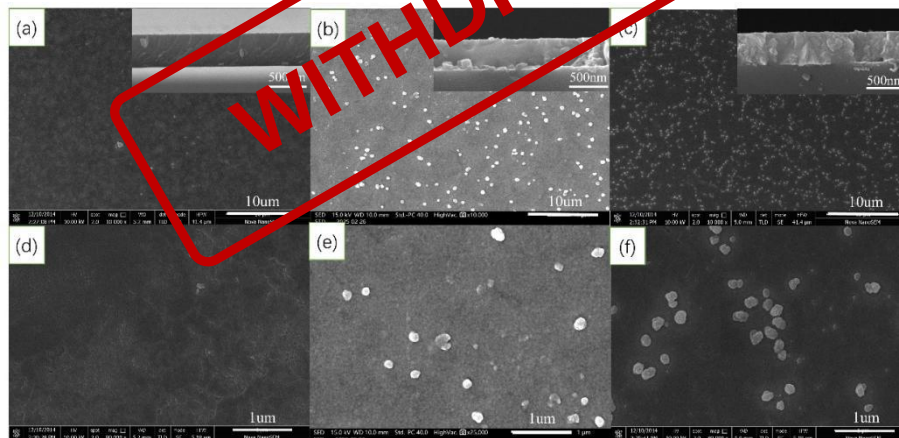


Figure 1: SEM images of gallium oxide films after annealing in (a) argon, (b) air and (c) oxygen atmospheres; the insets show cross-sectional views of the respective samples; (d), (e) and (f) are enlarged views of (a), (b) and (c) respectively.

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Oxygen Vacancy Segregation in Multi-Phase WO_{3-x} Thin Films Grown Under Tensile Strain

Joshua Feldon¹, Michal Kiaba¹, Laszlo Cline¹, Antoine Devinenti¹, and Jennifer Fowlie¹

¹ *Department of Materials Science and Engineering, Northwestern University*

WO₃ exhibits rich polymorphic phases and suboxides, garnering interest as an electronic material and possible high-temperature superconductor [1]. To further investigate these, stabilization of WO₃ single crystals in thin films is essential. To this end, pulsed laser deposition (PLD) is used to synthesize high crystalline quality, atomically flat, epitaxial WO₃ thin films on LaAlO₃ (LAO) substrates. This substrate induces a tensile strain to the film, and the effects have been studied using X-ray diffraction (XRD) [2]. It has also been discussed that this tensile strain applied to the film may decrease the formation energy of oxygen vacancies within the WO₃ lattice [3]. Soft chemical approaches are being investigated to transform WO₃ films to suboxide phases by introducing ordered oxygen vacancies to access its numerous suboxides (WO_{2.92}, WO_{2.9} (where metallic-to-insulator crossover behavior and filamentary evidence of superconductivity are observed), WO_{2.8}, WO_{2.76}, WO_{2.72}, WO_{2.66}, and WO_{2.63}). In this work, factors influencing oxygen vacancy ordering are investigated in thin films of WO₃ on LAO providing insight into the fundamental mechanisms governing the formation of tungsten suboxide phases.

It was observed in XRD measurements that by sufficient vacuum annealing of WO₃ thin films, the hexagonal phase of tungsten oxide is stabilized and that simultaneously, the crystalline quality of the as-grown monoclinic phase of WO₃ is improved, as is supported by the reappearance of Laue oscillations. It was further observed during an *in-situ* scanning electron microscope (SEM) heating experiment that the forming hexagonal phase was tungsten-rich compared to the lattice around it. Together, these results may suggest that the hexagonal phase preferentially absorbs oxygen vacancies, locally depleting the thin film lattice of the defects as well as the structural disorder and chemical changes that come with them. This defect-mediated interaction between the hexagonal and as-grown monoclinic phases of tungsten oxide not only has far-reaching effects for gas-sensing and catalysis applications, but also provides insight into the feasibility of growing scalable ordered tungsten suboxide devices.

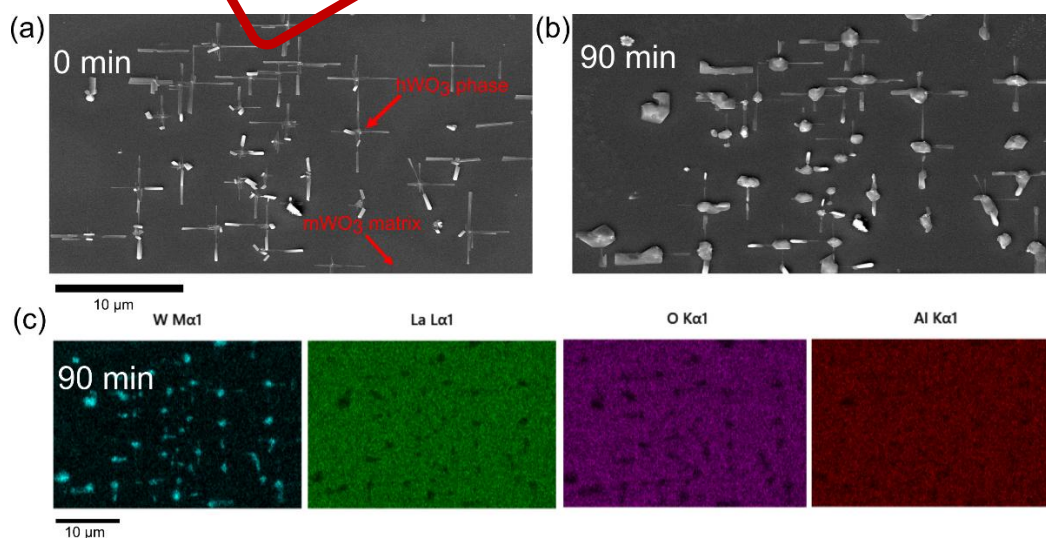


Figure 1. SEM secondary electron micrographs (a) just after hexagonal phase nucleation and (b) after 90 minutes of annealing as well as (c) energy-dispersive X-ray spectroscopy (EDS) elemental mapping micrographs.

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Optical Phonon Evolution in $\text{SrTiO}_{3-\delta}$ with Controlled Oxygen Vacancies

Dong Hyeon Kim¹, Jong Il Jeong¹, Dirk Wulferding^{2*}, Woo Seok Choi^{1*}

¹ Department of Physics, Sungkyunkwan University, Suwon 16419, Korea

² Department of Physics and Astronomy, Sejong University, Seoul 05006, Korea

SrTiO_3 (STO) is the prototypical perovskite oxide with a cubic structure at room temperature and exhibits myriads of intriguing physical properties. It undergoes structural phase transitions depending on temperature, doping, isotope substitution, or strain, for example, an antiferrodistortive (AFD) transition associated with the rotations of TiO_6 octahedra occurs at ~ 105 K. On the other hand, quantum fluctuation is known to suppress a ferroelectric (FE) transition involving Ti off-centering within the TiO_6 octahedra in pure STO without any external perturbations [1,2]. Oxygen vacancies (V_O) may break Ti-O bonds, inducing local distortion which alter existing or activate new phonon modes in STO.

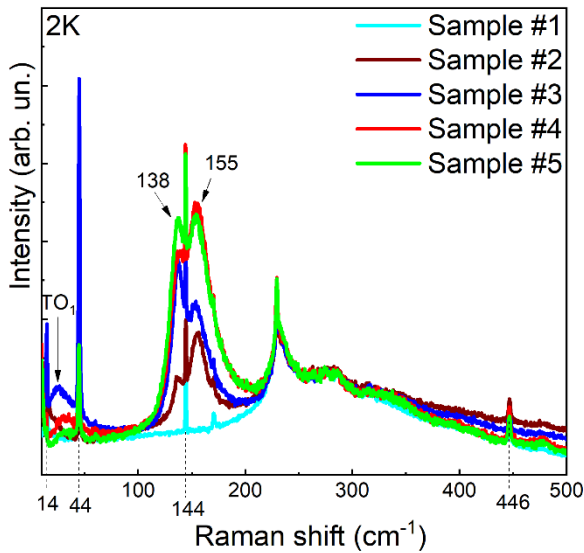


Figure 1: Normalized Raman spectra of $\text{SrTiO}_{3-\delta}$ at 2K without polarization

In this study, to examine the evolution of STO phonons with different V_O concentration, we employed five STO (100) samples, one pure STO and four V_O -induced STOs under different annealing conditions. Sample #1 is pure STO. Sample #2 was annealed at 800 K for 2.5 h. Sample #3, #4, and #5 were annealed at 850 K for 2.5h, 4h, and 6h, respectively. Annealing was conducted under 10^{-6} Torr. X-ray diffraction confirmed that all samples maintained the cubic structure at room temperature. We used Raman spectroscopy over the temperature range of 300 K to 2 K with and without polarization. Figure 1 shows representative Raman spectra for all the samples at 2 K. First, FE TO_1 soft phonon mode was observed at around $26 \sim 33\text{cm}^{-1}$. This mode

is Raman inactive in centrosymmetric STO but in the sample #3, #4, and #5, they become active as sufficient V_O break the local symmetry. As the V_O concentration increased, the TO_1 phonon frequency increased, while both its intensity and spectral weight decreased. This means that V_O hardened the TO_1 soft phonon mode and drove STO farther away from quantum criticality. Two additional peaks were observed in the oxygen-deficient samples around 138cm^{-1} and 155cm^{-1} , which are likely associated with new vibrational modes induced by V_O . Optical phonons related to AFD transition were also observed. The AFD soft mode splits into two phonon modes after transition which are E_g mode around 14cm^{-1} and A_{1g} mode around 44cm^{-1} at 2 K in all samples [3]. The frequencies of AFD soft phonons remained almost unchanged with increasing V_O concentration. Our results reveal the intrinsic properties of oxygen-deficient STO, showing the interplay between V_O , lattice dynamics, and FE instability.

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Controlling Oxygen Evolution Reaction in Ba-Doped LaNiO₃ Epitaxial Thin Films

Do-Hyun Kim¹, HyeonHui Na¹, Si Won Kim¹, Byungmin Sohn¹, Woo Seok Choi^{1,#}

¹ Department of Physics, Sungkyunkwan University, Suwon 16419, Republic of Korea

Developing highly efficient catalysts for the oxygen evolution reaction (OER), the rate-determining step of water splitting, is essential for sustainable carbon-free hydrogen production. 3d transition metal oxides are promising candidates for highly efficient OER catalysts owing to their excellent catalytic activity and substantially lower cost compared to noble metal-based catalysts such as Ru- and Ir-based materials. LaNiO₃ epitaxial thin films serve as a prominent model platform for 3d transition metal oxide catalyst. However, conventional strategies typically focus on independently modifying either the valence state [1] or the lattice strain [2]. In this study, we employ Ba doping as a strategy to simultaneously induce lattice expansion and diversify Ni oxidation states, thereby aiming for a synergistic modulation of OER performance.

We fabricated high-quality La_{1-x}Ba_xNiO₃ ($x = 0, 0.1, 0.2, 0.3$) epitaxial thin films on (001)-oriented Nb doped SrTiO₃ substrates using pulsed laser epitaxy. From X-ray diffraction patterns, systematic peak shifts with increasing Ba content reveal a progressive *c*-axis lattice expansion from 3.83 to 3.91 Å. X-ray absorption

spectroscopy and X-ray photoelectron spectroscopy analyses further reveal that Ba doping diversifies the Ni oxidation state from a Ni³⁺ configuration to a mixed Ni²⁺/Ni³⁺/Ni⁴⁺ distribution. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) indicate that the $x = 0.1$ and 0.2 films achieve the

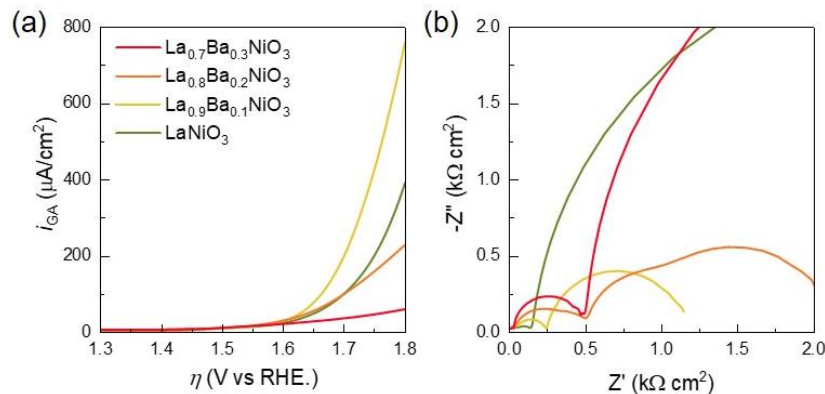


Figure 1. (a) CV results and (b) EIS results of La_{1-x}Ba_xNiO₃ thin films

highest OER performance in terms of onset overpotential within the series (Figure 1). We suggest that this superior activity is driven by the lattice expansion which shifts the O 2*p*-band center to favorably modulate the binding energy of oxygen intermediates, and the diversified Ni valence which shifts *e_g* orbital filling toward near-unity (≈ 1.2), thereby accelerating the OER kinetics. This study provides an effective design principle for advancing 3d transition metal oxide catalysts toward sustainable energy applications.

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Thermally quenched metastable metallic state in $V_{1-x}W_xO_2$

Hiroshi Oike¹, Yasunori Takahashi², Keisuke Shibuya³, Masaki Nakano⁴, Tatsuki Hanada²,
Motoaki Hirayama⁵, Hiroko Tokoro⁶, Fumitaka Kagawa^{5,7}

¹ *Research Center for Materials Nanoarchitectonics, National Institute for Materials Science*

² *Department of Applied Physics and Quantum-Phase Electronics Center, The University of Tokyo*

³ *National Institute of Advanced Industrial Science and Technology*

⁴ *Graduate School of Engineering and Science, Shibaura Institute of Technology*

⁵ *RIKEN Center for Emergent Matter Science (CEMS)*

⁶ *Department of Materials Science, University of Tsukuba*

⁷ *Department of Physics, Institute of Science Tokyo*

Thermal quenching can freeze atomic configurations before they reach the thermodynamic ground state by cooling a material faster than a phase transformation can proceed. This approach has long been used to access metastable states in a wide range of materials, including quenched alloys, structural glasses, and phase-change materials. However, whether a similar strategy can be applied to metal–insulator transitions, where the transformation is governed by collective electronic reorganization rather than long-range atomic diffusion, remains an open question.

In the present study, we focus on VO_2 , one of the most extensively studied metal–insulator transition systems, whose transition temperature can be widely varied by W substitution (Figure 1) [1]. Here we demonstrate that rapid cooling exceeding 10^9 K s⁻¹ induces a metastable metallic phase in $V_{1-x}W_xO_2$, although the equilibrium low-temperature phase is insulating. The metastable metallic state persists for more than 100 s, with temperature-dependent relaxation revealing a finite activation barrier that kinetically stabilizes the metallic phase.

Previous examples of metastable metallic phases have been mainly observed in geometrically frustrated triangular-lattice systems, where the degeneracy of charge configurations leads to a complex energy landscape with activation barriers separating metastable states [2]. In contrast, $V_{1-x}W_xO_2$ provides a non-triangular platform in which elemental substitution modifies the local structure and may introduce an activation barrier for the phase transition. This demonstrates that thermally quenched metastability can be achieved beyond geometrically frustrated materials and broadens the scope of thermal quenching as a strategy for controlling electronic phases.

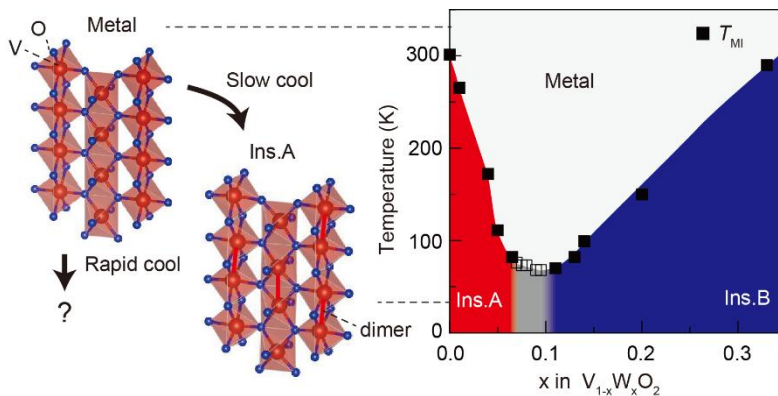


Figure 1: Phase diagram of $V_{1-x}W_xO_2$. The metal–insulator transition temperature T_{MI} is plotted as a function of W concentration x . The crystal structures of the equilibrium metallic and insulating phases are schematically shown for $x = 0$. The low-temperature state after thermal quenching is investigated in the present study.

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Stoichiometry control and structural tuning of CaIrO_3 thin films via plume dynamics

Alexandros Sarantopoulos¹, Kapil Nayak¹, David Gustin², Lee-Kang Huang¹, Omar Concepcion³, Joyce James¹, David Mueller¹, Felix Gunkel¹, Marjana Lezaic², Regina Dittmann¹.

¹ Peter Gruenberg Institute (PGI-7), *Forschungszentrum Juelich, Germany*

² Peter Gruenberg Institute (PGI-1), *Forschungszentrum Juelich, Germany*

³ Peter Gruenberg Institute (PGI-9), *Forschungszentrum Juelich, Germany*

Perovskite iridates have become a compelling ground for studying correlated and topological electronic phases, due to strong spin-orbit coupling of the Ir^{4+} ions which is energetically comparable to the on-site Coulomb interactions and electronic bandwidth. This interplay stabilizes a $J_{\text{eff}} = \frac{1}{2}$ ground state which can give rise to various emergent phenomena, from symmetry-protected topological semi-metals to spin-orbit assisted Mott insulators [1]. Within the iridate family, perovskite CaIrO_3 is an exciting prospect as a correlated Dirac semi-metal whose band degeneracy is protected by the non-symmorphic symmetry of its orthorhombic (Pbnm) lattice. Compared to the more extensively studied SrIrO_3 , the smaller Ca^{2+} ions drive larger IrO_6 octahedral rotations and a narrower bandwidth, enhancing the electron correlations by roughly 20% and placing CaIrO_3 substantially closer to the Mott transition [2]. The resulting state combines exceptionally high carrier mobility with very large magnetoresistance, establishing CaIrO_3 as an ideal system to investigate correlated Dirac phenomena. A crucial step in studying CaIrO_3 as thin films, is the stabilization of the perovskite phase and achieving cationic stoichiometry, which controls the IrO_6 octahedral tilting and hence the orthorhombic symmetry that protects the band degeneracy. Stoichiometry control is a major challenge in iridate growth, due to the volatile nature of the IrO_3 gas phase [3], yet systematic studies of the growth and stoichiometry of perovskite CaIrO_3 thin films remain scarce.

In this work we present our study on CaIrO_3 thin film growth by pulsed laser deposition, in which the cationic stoichiometry, the crystal structure, and the magneto-transport properties are tuned through the deposition parameters. We identify two off-stoichiometry mechanisms; the highest and lowest temperature regimes both yield Ir-deficient films, the former through enhanced IrO_3 volatility and the latter through insufficient oxidation. During this study, we systematically map the effects of pressure, temperature, and laser fluence on the magnetotransport properties, the crystal structure, and the cationic stoichiometry, revealing the interdependencies among these parameters, and identify growth conditions optimized for CaIrO_3 films under tensile strain. Suppressing the Ir volatility is a crucial step to control the cationic stoichiometry in the thin films but alone is not enough to produce high quality CaIrO_3 samples with the correct crystal symmetry. We have identified the importance of the laser plume dynamics and tuned them accordingly to obtain the desired properties. The growth window that increases Ir retention and stabilizes the Pbnm tilt order also recovers the intrinsic semi-metallic transport, establishing a direct chain connecting growth conditions, stoichiometry control, crystal structure, and transport properties, resulting in a practical route to engineering the properties of this correlated Dirac system.

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Low-Temperature Air-Annealing Effects on Electrical and Optical Properties of SrVO₃ Films

Hongming Zhang¹, Masamichi Negishi¹, Daichi Oka², Satoshi Sasaki¹,
and Tomoteru Fukumura^{1,3,4,*}

¹ *Department of Chemistry, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan*

² *Department of Chemistry, Graduate School of Science, Tokyo Metropolitan University, Hachioji 192-0397, Japan*

³ *Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

⁴ *Center for Science and Innovation in Spintronics, Tohoku University, Sendai 980-8577, Japan*

Transparent conducting oxides are key materials for transparent electronics. Among them, perovskite SrVO₃ has been extensively studied because of its high electrical conductivity without using rare metal elements[1]. At present, it is desired to improve the low carrier mobility and transmittance of SrVO₃. In this study, we report post-annealing effect on the structural, electrical, and optical properties of SrVO₃ thin films.

SrVO₃ thin films were grown on (LaAlO₃)_{0.3}(Sr₂TaAlO₆)_{0.7} (001) substrates by pulsed laser deposition method. Subsequently, the films were annealed at 150 or 200 °C for 24 h in air. From X-ray diffraction measurements, SrVO₃ in-plane peaks were not observed in the as-grown SrVO₃ (001) film without impurity phases, indicating low crystallinity along in-plane directions. After the annealing, an in-plane SrVO₃ 103 peak as well as 002 peak was observed, indicating the improved in-plane crystallinity to form the SrVO₃ epitaxial thin films. The non-annealed and annealed SrVO₃ films showed the metallic conduction as Fermi liquid (Fig. 1a). Although the electrical conductivity was not improved due to the large decrease in carrier concentration (Fig. 1(b)), the electron mobility was doubled reflecting the improved crystallinity (Fig. 1c). In addition, the absorption coefficient for infrared-red and ultraviolet regions was reduced by the annealing, particularly at 200 °C (Fig. 2). To summarize, low-temperature annealing in air is useful to improve both the carrier mobility and optical transparency of SrVO₃, though the additional control of the carrier density is necessary to improve the electrical conductivity.

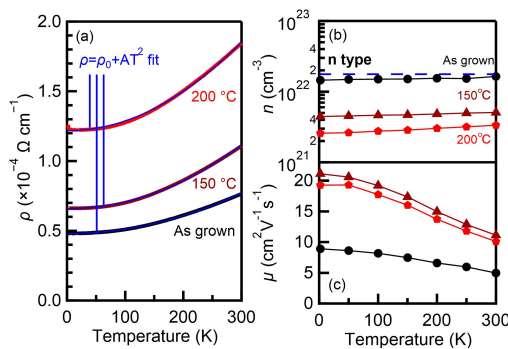


Figure 1. Temperature dependence of (a) resistivity, (b) electron density, and (c) electron mobility of the SrVO₃ films with different annealing temperatures.

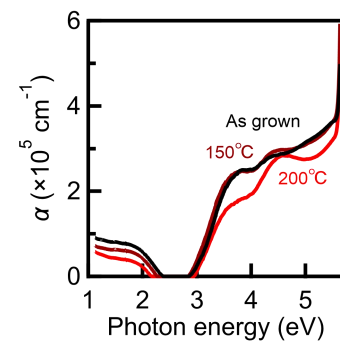


Figure 2. Optical absorption spectra of the SrVO₃ films with different annealing temperatures.

Growth and Characterization of Superconductor - Thermoelectric Interfaces

Chia-Hsuan Hsieh¹, Nicolas Bonmassar¹

¹*University of Stuttgart, Institute of Materials Science, Department for Materials Physics,
Heisenbergstr. 3, 70569 Stuttgart, Germany*

In this work, we combine oxide high- T_C superconductors with thermoelectric materials[1]. Thin films of $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$ (NBCO) are prepared by pulsed laser deposition (PLD)[2], following a deposition of LaNiO_3 to protect the NBCO from oxygen vacancy formation during the growth of the Bi-related thin films. A thermoelectric bilayer consisting of $\text{Bi}_{0.84}\text{Sb}_{0.16}$ and $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ is used to prove the possibility to combine high- T_C oxides with high performance thermoelectric materials[3]. The growth is monitored by *in situ* reflection high-energy electron diffraction (RHEED) confirming a 2D growth of all materials. The physical properties are investigated by electrical and magnetic measurements and the structure is determined by x-ray diffraction (XRD). A smooth surface was confirmed by atomic force microscopy (AFM) and the interface has been characterized at the atomic scale by transmission electron microscopy (TEM). Hence, we open a path for the combination of thermoelectric materials with a sharp interface to NBCO and combine the local cooling effect of thermoelectric materials with high- T_C superconductivity.

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Electronic Correlation Energy and Metal–Insulator Transition in VO₂/Nb:TiO₂ Heterostructures

Naoya Arihara¹, Taisei Yamada¹, Naohiro Egawa¹, Daisuke Shiga², Hiroshi Kumigashira², and
Tohru Higuchi¹

¹*Department of Applied Physics, Tokyo University of Science, Tokyo 125-8585, Japan*

² *Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan*

VO₂ exhibits a metal–insulator transition (MIT) near 340 K, accompanied by several orders of magnitude change in electrical resistance. This MIT is closely related not only to the structural phase transition but also to the electronic correlation of V 3*d* electrons, making VO₂ promising for resistance switching devices based on electronic-state and electric-double-layer control[1,2]. Conventional resistance switching in VO₂ has mainly been achieved by Joule heating and ion doping; however, these methods suffer from high power consumption and device degradation. In this study, VO₂/Nb:TiO₂ multilayer films were fabricated to investigate the effect of interfacial charge transfer on the electronic correlation and MIT properties of VO₂. Nb-doped TiO₂ (Nb:TiO₂) is a highly conductive n-type oxide that can induce charge transfer at the VO₂ interface, potentially modulating the electronic correlation and MIT characteristics of VO₂.

VO₂ single-layer and VO₂/Nb:TiO₂ multilayer films were fabricated on Al₂O₃(0001) substrates by RF magnetron sputtering. The thicknesses of the VO₂ layers were ~60 nm and ~140 nm. The crystal structures were evaluated by X-ray diffraction (XRD). The temperature dependence of electrical resistivity from 303 to 368 K was measured using the DC two-probe method. The electronic structures were investigated by X-ray photoelectron spectroscopy (XPS), X-ray absorption spectroscopy (XAS), and resonant photoemission spectroscopy (RPES) at Photon Factory BL-2A.

Fig.1 shows the XRD patterns of the VO₂ single-layer film and the VO₂/Nb:TiO₂ multilayer films. It was confirmed that the VO₂ films were *b*-axis oriented on Nb:TiO₂ layer with the same crystal structure. Fig.2 shows the temperature dependences of electrical resistivities. Compared with the VO₂ single-layer film, the VO₂/Nb:TiO₂ multilayer films exhibited a reduced resistance change and a shift of the MIT temperature toward higher temperatures. Fig.3 shows the electronic structures evaluated by RPES and XAS. At 295 K, the Lower Hubbard Band (LHB) and Upper Hubbard Band (UHB) were clearly observed. The electronic correlation energy U_{dd} , estimated from the energy separation between the LHB and UHB, was 2.55 eV for the heterostructure and 2.89 eV for the single-layer film, indicating that the electronic correlation was weakened in the heterostructure. These results suggest that charge transfer at the VO₂/Nb:TiO₂ interface relaxes the electronic correlation in VO₂, leading to changes in the MIT and resistance switching properties.

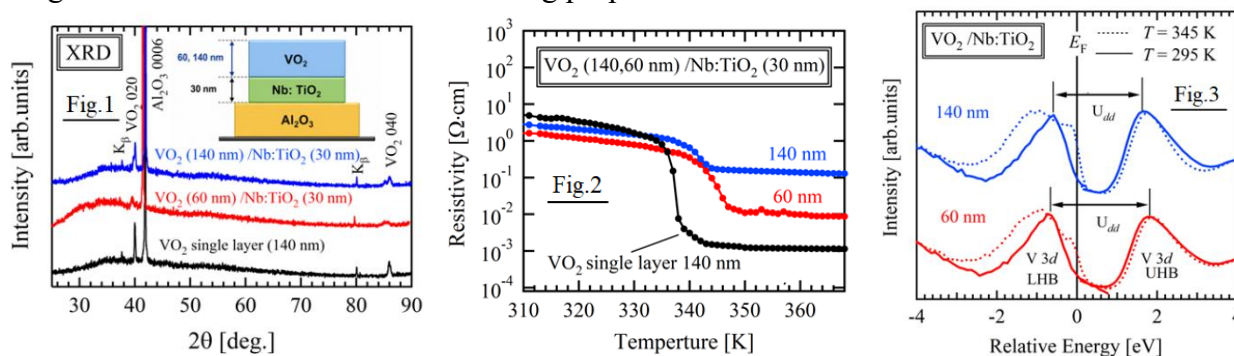


Fig.1: XRD patterns of VO₂ single-layer and VO₂/Nb:TiO₂ multilayer films. **Fig.2:** Temperature dependences of resistivities for VO₂ single-layer and VO₂/Nb:TiO₂ multilayer films. **Fig.3:** Estimation of U_{dd} from combined RPES and XAS spectra of VO₂/Nb:TiO₂ films

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Hierarchy of quantum corrections to resistivity of a γ -Al₂O₃/SrTiO₃ 2DEG

Jiwon Yang¹, Akira Endo¹, Daisuke Shiga^{2,3}, Hayato Konaka², Tomoaki Tanaka¹,
Masayuki Hashisaka¹, Hiroshi Kumigashira^{2,3}, and Mikk Lippmaa¹

¹ *Institute for Solid State Physics, The University of Tokyo, Kashiwa 277-8581, Japan*

² *Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Tohoku University, Sendai 980-8577, Japan*

³ *Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba 305-0801, Japan*

Unexpected logarithmic resistance upturns at low temperature have often been reported in SrTiO₃-based two-dimensional electron gases (2DEGs) and interpreted in terms of the Kondo scattering model. However, at low temperatures, where the phase coherence length of the electrons becomes comparable to the mean free path, quantum corrections such as weak (anti)localization and electron-electron interactions can also affect the resistance-temperature behavior. In particular, weak antilocalization has been reported in SrTiO₃-based heterostructures through magnetoresistance analysis, and its presence in these 2DEGs is well established.

In this presentation, we show that the low-temperature transport of the electron gas formed at the γ -Al₂O₃/SrTiO₃ interface is governed by a hierarchy of quantum corrections in addition to the Kondo contribution. Upon cooling, the resistance first exhibits a typical Kondo-like upturn, then decreases due to weak antilocalization, and finally vanishes at the superconducting transition. When superconductivity is suppressed by a magnetic field or by reducing the carrier density, a logarithmic correction from electron-electron interactions emerges at millikelvin temperatures, as shown in Figure 1.

Our results demonstrate that the low-temperature resistance cannot be explained by a Kondo-only interpretation and instead requires several quantum corrections to be taken into account.

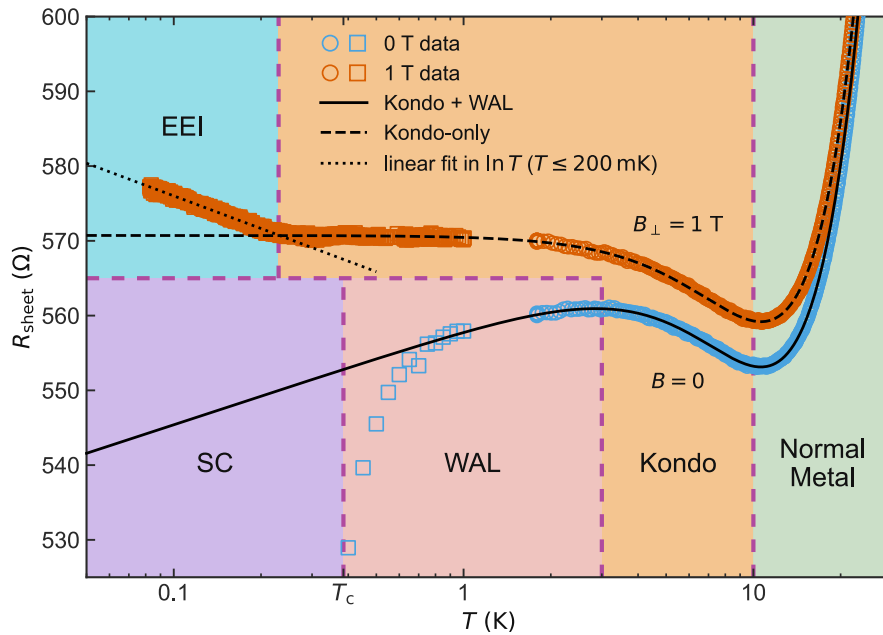


Figure 1: $R(T)$ at $B = 0$ (lower) and $B_{\perp} = 1$ T (upper) with fits. Shaded regions mark the dominant mechanism in each temperature range.

Two-gap superconductivity, unconventional pairing, and local field-effect control in KTaO_3 (111) 2DEGs

Marko Kuveždić¹, Vladimir Pirogov², Soumyajit Mandal², Hugo Witt^{1,2}, Isabella Boventer¹, Etienne Maréchal², Manuel Bibes¹, Nicolas Bergeal²

¹ *Laboratoire Albert Fert, CNRS, Thales, Université Paris-Saclay, 91767 Palaiseau, France*

² *Laboratoire de Physique et d'Étude des Matériaux, ESPCI Paris, Université PSL, CNRS, Sorbonne Université, 75005 Paris, France*

KTaO_3 -based two-dimensional electron gases are emerging as a distinctive platform for oxide superconductivity, combining relatively high superconducting transition temperatures, reaching up to 2.2 K [1], strong spin-orbit coupling [2], multiband electronic structure, and electrostatically tunable carrier density. These ingredients make KTaO_3 interfaces particularly attractive for investigating unconventional superconductivity and for the development of gate-defined superconducting devices.

We performed a combined spectroscopic and transport study of superconducting $\text{AlO}_x/\text{KTaO}_3$ (111) two-dimensional electron gases, revealing two-gap superconductivity, signatures consistent with unconventional pairing, and local field-effect control of the superconducting state. Tunnelling spectroscopy directly probes the superconducting density of states and reveals two distinct superconducting gaps, compatible with the multiband electronic structure observed by angle-resolved photoemission. Superconductivity in $\text{AlO}_x/\text{KTaO}_3$ (111) cannot be fully described by a simple single-band conventional picture, and a detailed analysis of the gap symmetry points towards unconventional pairing states, such as chiral d- or p-wave.

In addition, we demonstrate gate-controlled superconductivity using both global back gating and local top gating through a high- κ HfO_2 dielectric. Both control schemes allow continuous electrostatic tuning of the superconducting ground state and reversible switching across the superconductor-insulator transition. Superconducting field-effect transistor behaviour is demonstrated using dynamic modulation of the top gate, enabling reversible switching of the channel between superconducting and insulating states at modulation frequencies up to a few kilohertz, with the speed primarily limited by the external measurement circuitry. The demonstrated compatibility of KTaO_3 -based two-dimensional electron gases with top-gate device architectures provides a route towards locally reconfigurable superconducting circuits and gate-defined quantum devices.

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Control of the thermodynamic stability of protons in TiO₂/VO₂ heterostructures through TiO₂ cap-layer crystallinity

S. Fuji¹, L. Xie¹, T. Majima², R. Aso³, Y. Shimakawa¹, D. Kan^{1, 4}

¹ Institute for Chemical Research, Kyoto University, Japan

² Department of Nuclear Engineering, Kyoto University, Japan

³ Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University, Japan

⁴ Division of Applied Chemistry, Graduate School of Engineering, The University of Osaka, Japan

Proton insertion into transition-metal oxides enables nonvolatile and reversible control of their physical properties. We previously demonstrated that the TiO₂ cap layers on VO₂ thin films control proton migration through chemical potential mismatches at the TiO₂/VO₂ interface [1]. In this study, we show that TiO₂ cap-layer crystallinity is another key factor governing proton migration in TiO₂-capped VO₂ thin films.

TiO₂/VO₂ heterostructures were fabricated by pulsed laser deposition. VO₂ films were epitaxially grown on TiO₂(001) substrates, followed by TiO₂ cap-layer deposition. By depositing the TiO₂ cap layers at 400 °C and 350 °C, single-crystalline and polycrystalline-like TiO₂ cap layers were obtained, respectively, as confirmed by TEM observations. Figure 1 shows X-ray 2θ–θ diffraction patterns of as-grown TiO₂/VO₂ heterostructures and those after electrochemical proton insertion and subsequent air annealing at 100 °C.

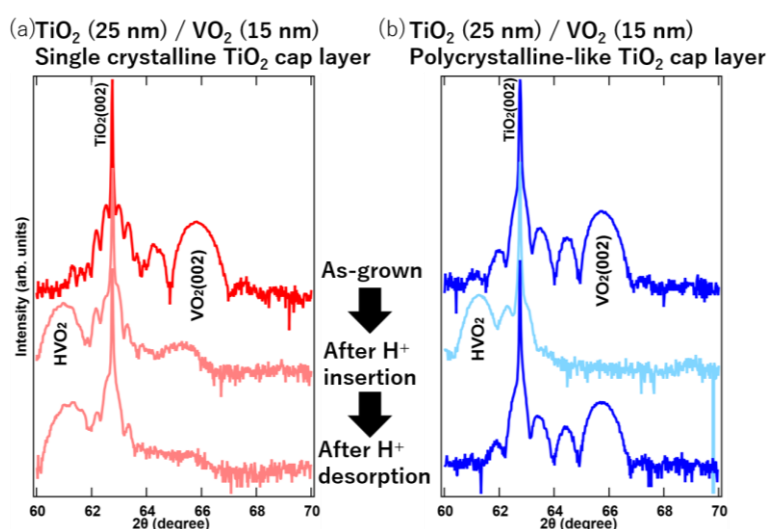


Figure 1. (a, b) X-ray 2θ/θ diffraction patterns of TiO₂(25 nm)/VO₂(15 nm) heterostructures with (a) the single-crystalline TiO₂ cap layer deposited at 400 °C and (b) a polycrystalline-like TiO₂ cap layer deposited at 350 °C, measured in the as-grown state, after proton insertion, and after air-annealing at 100 °C.

Regardless of the crystallinity of TiO₂ cap layers, TiO₂/VO₂ heterostructures after proton insertion exhibit new reflections that come from protonated VO₂, HVO₂, indicating successful proton insertion into VO₂ thin films through the TiO₂ cap layers. In contrast, the thermally-induced proton desorption behavior is found to depend on the cap-layer crystallinity significantly. With the polycrystalline-like TiO₂ cap layer, HVO₂ recovered to VO₂ after annealing, whereas with the single-crystalline TiO₂ cap layer, the HVO₂ phase was maintained as confirmed from the X-ray diffraction patterns. These results demonstrate that the cap-layer crystallinity is a key parameter for controlling the thermodynamic stability of protons in TiO₂-capped VO₂ thin films.

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Giant non-reciprocal transport from hexagonal Fermi-surface warping in a spin-polarized oxide Rashba electron system

Yu Chen¹, Janine Gückelhorn², Martando Rath¹, Gyanendra Singh², Emiliano Di Gennaro^{3,1},

Daniele Preziosi⁴, Gervasi Herranz², Daniela Stornaiuolo^{3, 1}, and Marco Salluzzo¹

¹ CNR-SPIN Complesso di Monte S. Angelo, via Cinthia, Napoli, 80126, Italy.

² Institute of Materials Science of Barcelona (ICMAB-CSIC), Campus UAB Bellaterra 08193, Spain

³ Department of Physics, University of Naples Federico II, via Cinthia, 80126 Napoli, Italy

⁴ Universite de Strasbourg, CNRS, IPCMS UMR 7504, F-67034 Strasbourg, France

Hexagonal Fermi-surface warping in Rashba two-dimensional electron systems (2DESs) can generate unconventional transport phenomena through complex spin-orbital textures. Here we report giant non-reciprocal transport in a spin-polarized (111) LaAlO₃/EuTiO₃/SrTiO₃ 2DES, where inversion, time-reversal, and rotational symmetries are simultaneously broken [1,2]. A large bilinear magnetoresistance and giant non-reciprocity emerge below 150 K, reaches a maximum near the ferromagnetic transition temperature ($T_c = 8$ K), and is suppressed in the ferromagnetic state. An empirical figure of merit of the nonreciprocity reaches approximately $10^{-7} T^{-1} A^{-1} cm^2$. To the best of our knowledge, this places our system one of the largest non-reciprocal transport responses reported to date. Its strong gate dependence reveals a close connection to the topology of the warped multi-orbital Fermi surface, while the low-magnetic field response uncovers competing contributions from magnetization and scattering with chiral magnons. Our findings identify magnetic Rashba oxide interfaces as a platform for engineering non-reciprocal transport through the interplay of spin-orbit coupling, electronic topology, and magnetism.

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Superconducting 2D Electron Gas on $\text{KTaO}_3(111)$ via Mg-Induced Surface Reduction

Chun Sum Brian Pang¹, Zhihao Chen^{2,3}, Bruce A. Davidson¹, Fengmiao Li¹, Mohamed Oudah¹, Peter C. Moen¹, Steef Smit¹, Cissy T. Suen^{1,4}, Simon Godin¹, Sergey A. Gorovikov⁵, Marta Zonno⁵, Pinder Dosanjh¹, Sergey Zhdanovich¹, Giorgio Levy¹, Matteo Michiardi¹, Alannah M. Hallas¹, George A. Sawatzky¹, Robert J. Green^{1,6}, Zhi Gang Cheng^{2,3}, Andrea Damascelli¹, Ke Zou¹

¹ *Quantum Matter Institute & Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia V6T 1Z4, Canada*

² *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

³ *School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China*

⁴ *Max Planck Institute for Solid State Research, 70569 Stuttgart, Germany*

⁵ *Canadian Light Source, Inc., Saskatoon, Saskatchewan S7N 2V3, Canada*

⁶ *Department of Physics & Engineering Physics, University of Saskatchewan, Saskatoon, Saskatchewan S7N 5E2, Canada*

Two-dimensional electron gases (2DEGs) at KTaO_3 surfaces have emerged as a promising system for investigating strong spin-orbit interactions, Rashba splitting, and superconductivity. In many existing approaches, however, carrier generation at KTaO_3 surfaces relies on the growth of chemically complex overlayers, which often masks the native electronic structure and hinders direct spectroscopic characterization. In this work, we present a simple route to realize a superconducting 2DEG on $\text{KTaO}_3(111)$ through Mg-induced surface reduction with molecular-beam epitaxy (MBE) [1]. Because Mg exhibits a very low sticking coefficient at elevated temperatures, the process yields only an ultrathin MgO overlayer ($<1-2$ monolayers), which remains sufficiently transparent to soft X-ray photoemission (XPS) and angle-resolved photoemission spectroscopy (ARPES). This enables direct access to both surface chemistry and electronic structure without any several-nanometer-thick capping layers. XPS indicates the emergence of reduced Ta species, while ARPES resolves a dispersive Ta 5d band with a bandwidth of approximately 150 meV and additional confinement-induced subbands. Electrical transport further reveals a superconducting transition below 0.7 K, with anomalous signatures consistent with a percolative evolution from locally coherent superconducting vortices toward a stripe-like electronic texture [2]. Our results introduce a straightforward platform for creating spectroscopically accessible oxide 2DEGs, while opening routes to systematically study orientation-dependent superconductivity in KTaO_3 and the role of disorder in 2D superconductors.

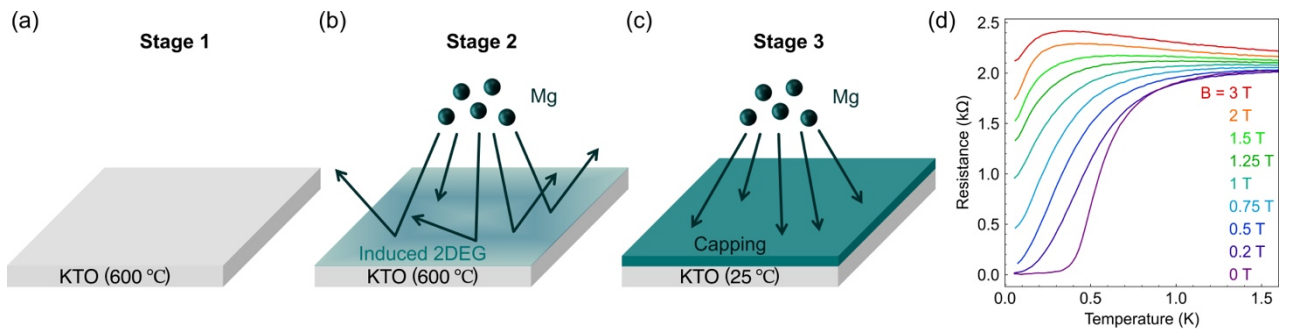


Figure 1: (a-c) Fabrication stages of the $\text{MgO}/\text{KTaO}_3(111)$ sample. (d) Low-temperature transport measurements of $\text{MgO}/\text{KTaO}_3(111)$ at varying out-of-plane magnetic fields, showing the superconducting transition. [1]

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Comparison of the Interfacial Charge Transfer in (100) and (111) $\text{LaMnO}_3/\text{SrMnO}_3$ Superlattices

Jun Ryeol Baek¹, Minho Lee¹, Heung-Sik Kim², Tuson Park¹ and Woo Seok Choi^{1*}

*Corresponding author: choiws@skku.edu

¹Department of Physics, Sungkyunkwan University, Suwon, Republic of Korea

²Department of Energy Engineering, Korea Institute of Energy Technology, Naju, Republic of Korea

LaMnO_3 (LMO) and SrMnO_3 (SMO) are A-type and G-type antiferromagnetic insulators, respectively [1]. However, recent theoretical and experimental studies have discovered ferromagnetic metallic (FM-M) states at their interfaces [1,2]. Superlattices (SLs) composed of LMO and SMO layers, with nominal stoichiometry comparable to $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, i.e., $(\text{LMO})_{2n}/(\text{SMO})_n$, exhibit thickness-dependent metal-insulator transition in addition to the interfacial FM-M phase [3]. The main origin of the emergent FM-M phase was proposed as mixed valence of Mn^{3+} and Mn^{4+} in both LMO and SMO layers by interfacial electronic charge transfer (CT) between the LMO and SMO.—

Recently, it was further calculated that (111)-oriented SLs remain strongly FM-M phase up to $n = 6$ compared to (100)-oriented SLs with FM-M phase only when $n = 1$ [2,3]. It is likely that the difference in the geometric symmetry influences electronic reconstruction at the interface. In particular, the Mn-O bond geometry of the (100)- and (111)-oriented heterointerfaces are distinct, and hence, the interfacial CT will differ [2]. The bond angle distortions will also be different, which may lead to a reduction of e_g orbital bandwidth for the case of (100)-orientation [4]. The larger e_g bandwidth in the (111)-oriented system facilitates higher magnetic moment by enhancing the double exchange. Such considerations may explain the theoretical prediction that (111)-oriented SLs exhibit wider CT region compared to (100)-oriented SLs. The comparison of (100)- and (111)-oriented SLs provides us with the control ability of electromagnetic properties by interface symmetry engineering.

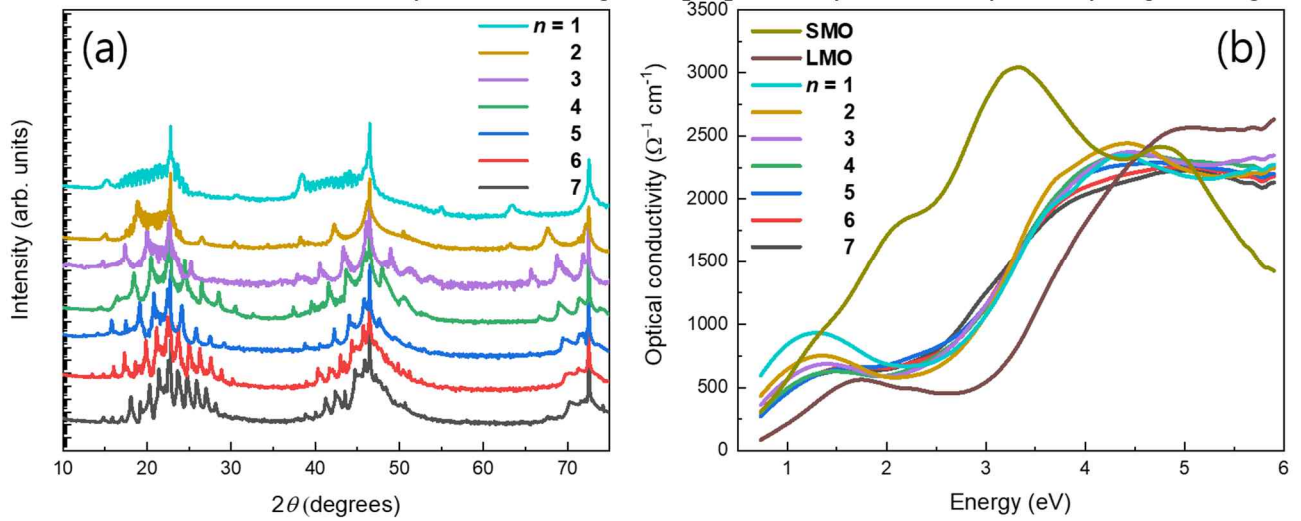


Figure 1. (a) XRD θ - 2θ of the $(\text{LMO})_{2n}/(\text{SMO})_n$ SLs. (b) Optical conductivity of the SMO, LMO and $(\text{LMO})_{2n}/(\text{SMO})_n$ SLs.

We have grown several $(\text{LMO})_{2n}/(\text{SMO})_n$ SLs first on (100)-oriented SrTiO_3 substrates (Figure 1(a)). Well-defined SLs satellite peaks are observed, indicating high crystalline quality and periodicity of the SLs. Low-energy optical conductivity is further shown to increase as n decreases, indicating more itinerant character, consistent with the theoretical prediction. We will also fabricate (111)-oriented SLs and measure the optical conductivity for comparing the thickness dependence of electric construction.

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Focused helium-ion beam engineering of gate-tunable tunnel barriers in $\text{LaAlO}_3/\text{SrTiO}_3$ interfaces

Yu Chen¹, Maria D'Antuono^{2,1}, Robin Hutt³, César Magén⁴, Edward Goldobin⁵, Dieter Koelle⁵, Reinhold Kleiner⁵, Marco Salluzzo¹, and Daniela Stornaiuolo²

¹*CNR-SPIN, Complesso di Monte Sant'Angelo–Via Cinthia, I-80126 Napoli, Italy*

²*University of Naples Federico II, I-80126 Napoli, Italy*

³*Physikalisches Institut, Center for Quantum Science (CQ) and LISA+, University of Tübingen, Auf der Morgenstelle 14, 72076 Tübingen, Germany*

⁴*Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC–Universidad de Zaragoza, 50009 Zaragoza, Spain*

⁵*Physikalisches Institut, University of Tübingen, Auf der Morgenstelle 14, 72076 Tübingen, Germany*

The $\text{LaAlO}_3/\text{SrTiO}_3$ (LAO/STO) interface hosts a high-mobility two-dimensional electron system exhibiting a remarkable range of emergent phenomena, including superconductivity, strong spin-orbit coupling and electrostatic tunability. These properties make LAO/STO an attractive platform for oxide nanoelectronics and quantum devices [1]. However, the realization of nanoscale device architectures remains challenging because of the buried nature of the conducting interface and the difficulty of locally tailoring its electronic properties with high spatial resolution.

Here we explore focused helium-ion beam (He-FIB) irradiation as a versatile technique for engineering nanoscale barriers in LAO/STO. He-FIB offers nanometric spatial resolution and has recently emerged as a powerful tool for the fabrication of functional oxide nanodevices, including superconducting Josephson junctions and SQUIDs [2]. We show that, in conducting LAO/STO interfaces, localized irradiation produces a controlled increase in resistance and enables the creation of narrow insulating regions embedded within the two-dimensional electron system without the need for complex multilayer processing [3]. Structural characterization reveals irradiation-induced lattice distortions confined to nanometric length scales, while transport measurements demonstrate the formation of gate-tunable barriers whose properties can be continuously controlled through electrostatic gating.

Low-temperature transport measurements reveal a crossover between different conduction regimes consistent with thermally activated transport, direct tunneling, and Fowler–Nordheim tunneling. Analysis of the gate-dependent current–voltage characteristics indicates that the effective barrier height can be continuously modulated by the applied gate voltage, providing access to a broad range of transport behaviors within a single device architecture. These results establish He-FIB irradiation as a promising route for the nanoscale functional engineering of oxide interfaces and open new opportunities for the development of tunnel devices, quantum point contacts, and more complex oxide nanoelectronic architectures.

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Ultralow-Temperature Magnetotransport at $\text{Dy}_2\text{Ti}_2\text{O}_7/(\text{Pb}_x\text{Sr}_{1-x})_2\text{Ru}_2\text{O}_{6.5}$ Heterointerface Structure

Yota Nakajima¹, Takahiro C. Fujita^{1,2}, Minoru Kawamura^{1,2}, Denis Maryenko², Masashi Kawasaki^{1,2}

¹ Dept. of Applied Physics, the University of Tokyo

² RIKEN CEMS

Pyrochlore oxide $\text{Dy}_2\text{Ti}_2\text{O}_7$ (DTO) is a canonical spin-ice compound. Strong Ising anisotropy confines each Dy^{3+} moment along its local $\langle 111 \rangle$ axis, and the effective magnetic interactions are geometrically frustrated on the pyrochlore lattice. As a result, the moments on each tetrahedron obey the “2-in/2-out” ice rule [1], while a magnetic field along [111] breaks this rule and flips the spins into the “3-in/1-out” configuration [2]. Both spin textures are non-coplanar, and their scalar spin chirality gives rise to an “emergent magnetic field” [3]. However, in insulating DTO, this emergent field is inaccessible to conventional electrical transport probes.

One strategy to overcome this limitation is to couple DTO to a conducting layer, allowing carriers in the adjacent layer to sense the emergent magnetic field at the heterointerface. This approach has revealed intriguing magnetotransport phenomena associated with the spin-flip transition in DTO, including anomalous magnetoresistance in $\text{DTO}/\text{Bi}_2\text{Ir}_2\text{O}_7$ [4] and the topological Hall effect in $\text{DTO}/\text{Bi}_2\text{Rh}_2\text{O}_7$ [5]. Here, we focus on $(\text{Pb}_x\text{Sr}_{1-x})_2\text{Ru}_2\text{O}_{6.5}$ (PSRO) as an alternative conducting oxide. In contrast to these previously studied heterostructures, DTO/PSRO can be grown entirely in situ and retains metallic conduction down to the lowest temperatures, even in the ultrathin-film limit (~ 3 nm). Leveraging these advantages, we previously demonstrated thickness-dependent interfacial transport in DTO/PSRO heterostructures down to 2 K [6]. However, this temperature remains relatively high for spin-ice physics, since the spin dynamics of DTO freeze out below approximately 1 K [7].

In this work, we examine the ultralow-temperature magnetotransport of this heterostructure down to ~ 50 mK. Upon cooling to this temperature, the magnetoresistance ratio (MRR) changes markedly. At 50 mK it exhibits anomalous structures and hysteresis (Figs. 1 and 2) that are absent at 2 K, reflecting the change from the 2-in/2-out to the 3-in/1-out states [2]. These results indicate that the spin-ice behavior of the DTO layer with minimal thermal fluctuations is imprinted onto the metallic PSRO layer.

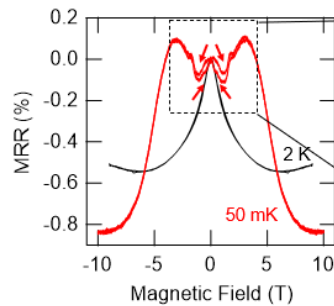


Fig. 1 MRR at 2 K and ~ 50 mK

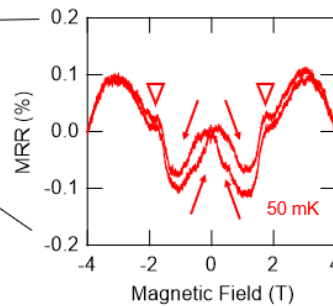


Fig. 2 clear anomaly and hysteresis at ~ 50 mK

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Real-space mapping of orbital quantum geometry at oxide interfaces

Chang Jae Roh¹, Maria Teresa Mercaldo², Giacomo Sala¹, Xi Wang^{4,5}, Eylon Persky^{4,5}, Thierry. C. van Thiel⁶, Stefano Gariglio¹, Beena Kalisky^{4,5}, Carmine Ortix^{2,3}, and Andrea D. Caviglia^{1,*}

¹ *Department of Quantum Matter Physics, University of Geneva, Geneva, Switzerland*

² *Dipartimento di Fisica ‘E. R. Caianiello’, Università di Salerno, Fisciano, Italy*

³ *CNR-SPIN c/o Università di Salerno, Fisciano, Italy*

⁴ *Department of Physics, Bar-Ilan University, Israel*

⁵ *Institute of Nanotechnology and Advanced Materials, Bar-Ilan University, Israel*

⁶ *Kavli Institute of Nanoscience, Delft University of Technology, Delft, Netherlands*

In quantum materials, the geometry and topology of electronic wave functions underpin emergent electrodynamic phenomena[1]. For instance, Berry curvature governs nonlinear electronic transport as constrained by crystallographic symmetry. Interfaces in oxide heterostructures allow the synthetic breaking of inversion symmetry, which enables the orbital Rashba effect that reshapes the orbital electronic structure, thereby inducing momentum-space-sourced Berry curvature[2,3]. Despite its importance, the direct connection between crystallographic symmetry and orbital quantum geometry remains uncharted.

In this work, we map the spatial distribution of orbital quantum-geometric responses in two-dimensional electron systems at (111)-oriented LaAlO₃/SrTiO₃ interfaces. To link local crystallographic symmetry with nonlinear electronic transport, we combine optical second-harmonic generation, scanning SQUID microscopy, and symmetry-based modeling. We characterize the interface symmetry across the antiferrodistortive transition from point group C_{3v} to C_s, producing equivalent domains with a polar plane that we image optically. The domain structure defines local orbital textures that allow a Berry curvature dipole in the conducting interfaces. Local second-harmonic current-density maps reveal a nonlinear electronic response with a spatial structure defined by the polar domain configuration, whose amplitude is governed by the orientation of the remaining mirror plane. These results identify interface symmetry as a real-space control parameter for orbital quantum geometry, providing a route to manipulate Berry-curvature-driven responses in quantum materials.

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From Chromium Telluride to Oxide heterostructures: Ageing and Oxidation of Chromium Tellurides

Pia Henning¹, Anna Tschesche¹, Tobias Meyer¹, Jasnamol Palakkal¹

¹ *Institute for Materials Physics, Georg-August-University Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany*

Transition metal dichalcogenides (TMDCs) have attracted significant attention due to their multifunctional properties and potential for electronic and spintronic applications [1]. Among them, chromium tellurides, $\text{Cr}_{(1+\delta)}\text{Te}_2$, are particularly promising because their tunable magnetic properties are influenced by Cr self-intercalation. Depending on δ , they exhibit ferromagnetism, with Curie temperature (T_C) varying from 125 K ($\delta = 0$) to 325 K ($\delta = 1/3$), with switchable perpendicular magnetic anisotropy, originating from modifications of the exchange interactions between the CrTe_2 layers [2,3].

Despite their technological potential, $\text{Cr}_{(1+\delta)}\text{Te}_2$ compounds exhibit limited air stability, which restricts practical applications [4]. Understanding the oxidation and ageing mechanisms is therefore essential and may further enable the design of multifunctional oxide/TMDC heterostructures through controlled oxidation. For example, $\text{Te/Cr}_2\text{O}_3/\text{CrTe}_6$ heterostructures exhibit ultra-high exchange bias fields, highlighting the potential of oxidation-engineered chromium tellurides for spintronic devices [5].

In this work, we investigated the oxidation behavior of $\text{Cr}_{(1+\delta)}\text{Te}_2$ thin films deposited on Al_2O_3 (0001) substrates by hybrid pulsed laser deposition (hPLD) [2]. In this approach, Cr is supplied via conventional pulsed laser deposition, while a valved cracker effusion cell provides a controlled Te flux, enabling precise adjustment of the intercalation level δ and excellent control over thin-film growth. Detailed structural, magnetic and magnetotransport properties of CrTe_2 and Cr_2Te_3 thin films were performed [2]. The samples were subsequently stored under ambient conditions and re-examined for surface oxidation using Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and cross-sectional energy-dispersive X-ray spectroscopy (EDX) after 1, 1.5, and 2.5 years of air exposure, and confirmed oxide formation.

The morphology and chemical composition of the oxide layers, as well as the underlying ageing mechanisms, were investigated using cross-sectional EDX, revealing significant Te depletion within the oxidized surface region. Furthermore, the influence of surface oxidation on the magnetic properties of the films was evaluated. A reduction in both the magnetization and T_C was observed only after prolonged air exposure exceeding one year, indicating a relatively slow degradation process despite the pronounced surface oxidation. These results provide valuable insights into the long-term ageing behavior and oxidation mechanisms of $\text{Cr}_{(1+\delta)}\text{Te}_2$ thin films, establishing a foundation for the future design and utilization of oxide/TMDC heterostructures derived from controlled oxidation processes [2,6].

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Electronic structure of Mo-doped SrRuO₃ probed by photoemission spectroscopy

Hyeonhee Na¹, Rahma Dhani Prasetyawati¹, Jeonghwan Oh¹, Se young Park², Woo Seok Choi¹,
Byungmin Sohn¹

¹ *Department of Physics, Sungkyunkwan University, Suwon 16419, Republic of Korea*

² *Department of Physics, Soongsil University, Seoul 06978, Republic of Korea*

Among functional oxides, SrRuO₃ (SRO) hosts a wide range of phenomena associated with magnetism and electron correlation, such as itinerant ferromagnetism, two-dimensional van Hove singularities, and the anomalous Hall effect (AHE). Because these are sensitive to band filling, controlling the carrier density is a powerful route to investigating them systematically. Mo substitution was recently shown to enhance the AHE and raise the Curie temperature of SRO films [1], but the underlying electronic structure has not been measured. Here, X-ray and angle-resolved photoemission spectroscopy show that Mo enters in mixed 5+/6+ states with a corresponding growth of Ru³⁺, so that each Mo donates one to two electrons and the filling change exceeds the nominal Mo concentration. The Fermi surface expands accordingly, following a rigid-band shift. Mo substitution thus can provide a quantitatively predictable route to tuning the band filling of SRO, and the electronic-structure basis for its correlation-driven magnetism and transport, including the AHE.

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Realization of an octahedral rotation-free surface in SrRuO₃

Jinju Lee¹, Jinyoung Kim², Changyoung Kim^{*2}, Minjae Kim^{†3,4}, and Byungmin Sohn^{‡1}

¹ *Department of Physics, Sungkyunkwan University, Suwon 16419, Korea*

² *Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea*

³ *School of Computational Sciences, Korea Institute for Advanced Study, Seoul 02455, Korea*

⁴ *Department of Semiconductor Science & Technology, Jeonbuk National University, Jeonju, 54896, Korea*

**Email: Changyoung@snu.ac.kr, †Email: mjkim1985@jbnu.ac.kr, ‡Email: bsohn@skku.edu*

Abstract

High density of states (DOS) near Fermi level enhances electronic instabilities in correlated materials, often leading to exotic quantum phases and structural distortions. In SrRuO₃, octahedral rotations stabilize the lattice by inducing t_{2g} - e_g hybridization, which opens a gap and reduces the high DOS associated with van Hove singularities. Here, using a combination of angle-resolved photoemission spectroscopy (ARPES), low-energy electron diffraction (LEED), and density functional theory (DFT) calculations, we show that tuning the surface chemical potential suppresses this stabilization mechanism and drives the near-surface region toward a rotation-free structural regime. Electron doping shifts the Fermi level away from the hybridized states, thereby reducing the hybridization-driven energy gain that favors the rotated structure. As a result, the RuO₆ octahedra relaxes into a rotation-free configuration. Our findings demonstrate that electronic-structure control can directly modulate lattice symmetry and establish a new route for engineering surface-driven quantum phases in correlated oxides.