Advanced Microelectronic Materials and Devices Mini-Symposium
Room 318 - Session AM+EM+TF-ThM

Ferroelectric Thin Films

Moderators: John F. Conley, Jr., Oregon State University, **Daniel Pennachio**, Naval Research Laboratory

8:00am **AM+EM+TF-ThM-1 Processing and Properties of Ferroelectric Wurtzite Thin Films**, ***Jon-Paul Maria,*** Penn State University **INVITED**

Ferroelectricity in wurtzite-based crystals was observed in 2019 and immediately introduced exciting opportunities to explore and discover new structure-property relationships in novel formulation spaces. These observations lead one to speculate that ferroelectricity might be found much more broadly, even “everywhere”, by introducing the appropriate disorder in a variety of hosts.

The presentation will begin with a brief history of ferroelectricity with specific attention to the last 10 years where this important property was discovered in new oxide and nitride crystals. The remaining content will focus on the structure-process-property relationships in the B-substituted AlN and Mg-substituted ZnO wurtzite systems. Materials can be prepared between 100 °C and 350 °C with very little difference in electrical properties. In the best cases, capacitors can be prepared down to 10 nm thickness while still exhibiting ferroelectric switching. Below 25 nm, however, leakage current becomes problematic during low frequency hysteresis measurements. Challenges to thickness scaling will be discussed with attention to the origins of property dependencies on structure , defects, and microstructure with film thickness. The presentation will also include examples where proximity effects in layered ZnO/$Zn_{1-x}Mg_xO$, AlN/$Al_{1-x}B_xN$, and $Zn_{1-x}Mg_xO$/AlN heterostructures can induce switching in pure ZnO and AlN layers, with opportunities for reducing net coercive voltage values.

8:30am **AM+EM+TF-ThM-3 Interfacial Control and Property Enhancement Through Proximity Ferroelectricity**, ***Ian Mercer,*** *William Prudnick, Saeed Almishal, Pochun Hsieh, Stanislav Udovenko, Darren Pagan, Venkatraman Gopalan, Jon-Paul Maria,* Penn State University

Ferroelectric layers can induce switching in neighboring polar non-ferroelectric layers, this phenomenon is referred to as proximity ferroelectricity. Prior studies have only focused on bi- and trilayer geometries, leaving room to further study on interface effects. Here, we explore interfacial and surface contributions more broadly, including, for example, the density effects in AlN/$Al_{1-x}B_xN$ and AlN/$Al_{1-x}Sc_xN$ superlattices grown using reactive magnetron sputtering. In addition, we examine compositionally graded structures that smear the same chemical contrast into an interface-less continuum. To complement this understanding, we also compare materials whose surfaces are plasma modified to create a partnering interface that can also influence switching. Thin film morphology and structural quality is probed using atomic force microscopy (AFM) and x-ray diffraction (XRD), revealing smooth films with RMS roughness below 1 nm in all cases. For superlattices, distinct satellite peaks indicating sharp, periodic interfaces are present. Deep UV photoluminescence (PL) measurements surprisingly reveal evolution with increasing interface density. By examining coercive field, current transport, and switching kinetics, we find that the internal interfaces act as barriers to both domain propagation and leakage transport. We also find grading $Al_{1-x}Sc_xN$ from x = 0.38 to x = 0 through the film thickness produces a substantial increase in coercive field, with some structures exhibiting strong insensitivity to Sc composition. These results establish interface, compositional grading, and surface processing – well-established opportunities in III-nitride alloys – as new degrees of freedom in wurtzite ferroelectric materials, demonstrating that engineered surfaces and gradients can strongly modify bulk switching behavior beyond conventional composition control. This transition towards interface engineering allows finer control over coercive field, domain structure, and leakage current barriers, ultimately enabling surfaces to enhance navigation through property space.

8:45am **AM+EM+TF-ThM-4 Unveiling the Hidden Role of Interfacial Layer in Ta/HZO Ferroelectric Tunnel Junctions Band-Profile and Tunneling-Electroresistance Engineering**, ***Sanghyun Lee,*** University of Kentucky*; Kent Price,* Morehead State University*; M. David Henry, Samantha Jaszewski,* Sandia National Laboratories, USA*; Alexander Blevins,* University of Kentucky

Ferroelectric tunnel junctions (FTJs) are promising two-terminal devices for neuromorphic computing, providing non-volatile, analog-tunable conductance via polarization-controlled tunneling. Their compact metal–ferroelectric–metal structure, low programming energy, and compatibility with CMOS processes make them suitable as synaptic elements for computing-in-memory. Hafnium oxide-based systems, such as $Hf_{0.5}Zr_{0.5}O_2$ (HZO), are especially notable for maintaining robust ferroelectricity at thicknesses below 10 nm, which supports scalable crossbar arrays for high-density neuromorphic computing.

The electrical performance of metal/HZO/metal FTJs is significantly influenced by interfacial chemistry; however, previous research efforts have consistently assumed an ideal, abrupt metal/HZO interface. In particular, Ta/HZO or Ti/HZO devices tend to form the unavoidable sub-stoichiometric TaO_x or TiOx layer due to tantalum's oxygen-scavenging properties. To bridge this research gap, we have investigated the interface layer and their material and device characteristics to unveil key physical mechanism. Utilizing in-house MATLAB modeling suites with Sentaurus TCAD simulators, the impact of this interlayer on band profile, tunneling pathways, and the resulting tunneling electroresistance (TER) is quantitatively assessed.

After fabricating and validating Ta/HZO (6 nm)/TaN devices, we characterized FTJs and developed a graded TaO_x interlayer device model with thicknesses from 0.3 to 1.5 nm and various permittivity values. Our model incorporates realistic oxygen vacancy concentrations (10^{19}–10^{21} cm^{-3}) and trap levels 0.5–1.2 eV below the HZO conduction band, which agrees well with experimental data. By integrating ferroelectric polarization, band bending, vacancy charge balance, the simulations represent the complete electrostatic and defect-mediated conduction mechanism at the Ta/HZO interface.

Our preliminary results uncover distinct regimes in which TaO_x either enhances or degrades FTJ performance. Thin, moderately defective layers (approximately 0.5 to 0.8 nm, oxygen vacancies around 10^{19} cm^{-3}) create a beneficial asymmetric barrier that increases TER by a factor of 3 to 6 compared to an ideal Ta contact, while maintaining read current. In contrast, thicker or highly oxygen-deficient TaO_x layers function as parasitic resistive elements, reducing low-resistance state current by more than an order of magnitude and generating significant internal bias fields. Simulated internal bias fields and coercive voltage shifts align with experimentally observed imprint trends, supporting the validity of the model.

9:00am **AM+EM+TF-ThM-5 Processing Window and Property Enhancement via Tellurium Co-Doping in Sputtered Ferroelectric $Zn_{1-x}(Mg, Te)_xO$ Thin Films**, ***William Prudnick,*** *Ian Mercer,* Pennsylvania State University*; Ece Günay,* Carnegie Mellon University*; Po Chun Hsieh, Saeed Almishal,* Pennsylvania State University*; Elizabeth Dickey,* Carnegie Mellon University*; Venkatraman Gopalan, Jon-Paul Maria,* Pennsylvania State University

Wurtzite ferroelectric materials show promise for application in non-volatile random-access memory (RAM). $Zn_{1-x}Mg_xO$ (ZMO), a wurtzite material first reported to be ferroelectric in 2021, shows significantly higher polarization retentions than perovskite ferroelectrics, similar to $Al_{1-x}Sc_xN$ and $Al_{1-x}B_xN$ but with markedly lower coercive fields compared to other wurtzite ferroelectrics (~2-3 MV/cm vs. ~4-6 MV/cm). However, leakage current, imprint, and intolerance for elevated growth temperatures remain persistent hinderances. Herein, we report tellurium incorporation within ZMO via reactive RF co-sputtering. $Zn_{1-x}(Mg, Te)_xO$ (ZMTO) films maintain consistently low coercive fields (~2.4 MV/cm) even at elevated growth temperatures (up to 350° C with no diminishment in ferroelectric properties) while simultaneously decreasing imprint. X-ray diffraction (XRD) data indicate highly c-oriented films with improved out-of-plane crystallinity. Omega rocking curves show reduction in mosaicity compared to previously reported values (< 2° FWHM vs. ~4° FWHM). X-ray reflectometry (XRR) data demonstrate low rms roughness values consistently below 1.5 nm. Transmission electron microscopy (TEM) data indicate columnar growth and planar defects such as stacking faults consistent with previous reports. Selected-area electron diffraction (SAED) supports wurtzite phase uniformity. X-ray photoelectron spectroscopy (XPS)

indicate tellurium is in the +4 oxidation state. Fourier transform infrared (FTIR) and Raman spectroscopies show tellurium with a 4-coordinate geometry indicative of Zn-site occupancy. We highlight photoluminescence spectroscopy (PL) data and expound upon dopant and temperature dependence of the defect landscape. We emphasize the underlying defect chemistry and doping strategies to overcome challenges in synthesis as they relate to material properties and structure enhancements.

9:15am **AM+EM+TF-ThM-6 Structurally Compatible Al-doped ZnO Bottom Electrode for Wurtzite Ferroelectric Mg-substituted ZnO Thin Films**, ***Sharon Yang***, The Pennsylvania State University; *Ece Gunnay,* Carnegie Mellon University; *William Prudnick,* The Pennsylvania State University; *Elizabeth Dickey,* Carnegie Mellon University; *Jon-Paul Maria,* The Pennsylvania State University

Epitaxial wurtzite heterostructures are prepared to eliminate, to the extent possible, structure defects and microstructure that may, in principle, interfere with or limit their ferroelectric properties and performance. In so doing, we attempt to achieve and understand the true intrinsic response. To do so, we begin with epitaxial c-axis oriented Al-doped ZnO (AZO) bottom electrodes grown on (0006)-Al_2O_3 via dual-cathode reactive RF magnetron co-sputtering. X-ray diffraction (XRD) indicates a single-phase c-axis oriented growth with x-ray omega and two-theta peak widths, electrical resistivity, and surface roughness values that all decrease with increasing substrate temperature and aluminum cathode power. The optimal AZO thin films exhibit an omega FWHM of 0.04° and surface roughness of 900 pm RMS, and a peak Hall mobility of 47 $cm^2/V{\cdot}s$, a Hall carrier density of 1.3E20 cm^{-3}, a minimum Hall resistivity of 1E-4 Ω·cm, and a STEM-EDS confirmed a composition of 1.7 at.% Al and uniform thickness of 43 nm. Infrared spectroscopic ellipsometry as function of Al content demonstrates tunable epsilon-near-zero behavior where the zero-permittivity wavelength ranged between 1970 and 3170 wavenumbers for 4.1E19 cm^{-3} and 1.3E20 cm^{-3} carrier concentrations respectively. Subsequently, $Zn_{0{\cdot}78}Mg_{0{\cdot}22}O$ (ZMO) films were deposited at room temperature, XRD indicating a strong c-axis growth of ZMO on AZO with FWHM of 0.86°. High-resolution TEM and STEM-EDS analysis revealed chemically abrupt ZMO/AZO interfaces. The heterostructure displays robust ferroelectric switching with a remanent polarization exceeding 88 $\mu C{\cdot}cm^{-2}$ and a coercive field below 2.8 $MV{\cdot}cm^{-1}$. Compared to previously reported ZMO films grown on non-epitaxial Pt bottom electrodes, these epitaxial oxide heterostructures exhibit comparable ferroelectric switching behavior while providing substantially improved structural compatibility and crystallographic coherence throughout the stack. These stacks are additionally attractive because they remain transparent throughout the visible spectral range, and these findings establish epitaxial AZO as a viable, highly compatible oxide bottom electrode platform for high-performance wurtzite ferroelectric thin films.

9:30am **AM+EM+TF-ThM-7 Stabilization of Antipolar Pbca Phase in Hafnium Zirconium Oxide via Aluminum Substitution**, ***Yong Kyu Choi***[1], University of Virginia, USA; *MinChul Kang,* Ames National Laboratory; *Tinase Ale,, Nicolas Lam, Andrea Watson, Kory Burns, Stephen McDonnell, Lin Zhou, Jon Ihlefeld,* University of Virginia, USA

Hafnium zirconium oxide (HZO) shows significant potential for non-volatile memory applications due to its ferroelectric properties. However, practical limitations such as insufficient endurance remain a significant challenging prior to commercialization. One promising approach to overcome this limitation is the implementation of antiferroelectric (AFE) behavior in HZO-based materials. Previous studies have demonstrated that cation substitution in HZO can induce AFE-like behavior within certain substitution concentration ranges. In Zr-rich HZO systems, this AFE response has commonly been attributed to the tetragonal phase. However, recent reports indicate that the observed AFE behavior may instead originate from the antipolar orthorhombic (*Pbca*) phase. This presentation discusses how varying an aluminum substitution concentration influences the phase stability and electrical properties of HZO thin films. Grazing-incidence X-ray diffraction, Fourier transform infrared spectroscopy, and scanning transmission electron microscopy are employed to investigate the structural transformation and AFE behavior of Al-substituted HZO capacitors. The results demonstrate that low aluminum substitution concentrations, such as 8:1 and 6:1 Al:HZO ratios, exhibit AFE behavior associated with stabilization of the antipolar orthorhombic *Pbca* phase.

9:45am **AM+EM+TF-ThM-8 Morphotropic Phase Boundary Like Dielectric Enhancement in ALD Deposited ZrO_2-Based Thin Film**, ***Myeongchan Ko***, Korea Advanced Institute of Science and Technology (KAIST), Republic of Korea; *Beatriz Noheda,* University of Groningen, Netherlands; *Kyung Min Kim,* Korea Advanced Institute of Science and Technology (KAIST), Republic of Korea

Ferroelectric HfO_2–ZrO_2-based thin films have emerged as promising high-permittivity dielectrics, yet the physical origin of their dielectric enhancement remains debated. Previous studies on HZO have reported dielectric enhancement that strongly depends on temperature, electrical history, and preceding bias conditions, suggesting substantial contributions from field-induced structural transitions. Here, we demonstrate a dielectric enhancement in Ta-doped ZrO_2 thin films, which exhibit a high dielectric constant of 65, without high-voltage preconditioning while maintaining a nearly temperature-independent response. Atomic-resolution imaging reveals a gradual structural evolution from the tetragonal phase through an intermediate transition region to the orthorhombic phase, accompanied by an increase in the projected Zr–Zr spacing, providing direct evidence of tensile-strain evolution at the atomic scale. Synchrotron diffraction further confirms the persistent coexistence of tetragonal and orthorhombic phases and reveals a thermally driven redistribution of orthorhombic crystallographic orientations rather than a complete phase transformation. Together, these structural observations indicate a strain-softened, shallow energy landscape in which closely competing structural states remain readily accessible under thermal and electrical perturbations, thereby enhancing dielectric susceptibility without relying predominantly on a field-induced phase transition. These results reveal that large dielectric susceptibility in fluorite oxides does not necessarily require field-driven phase transformation but can instead emerge from a strain-softened landscape of competing polymorphs. In contrast to the transition-mediated dielectric enhancement in HZO from many previously reported studies where high voltage sweep is required to obtain high dielectric constant, Ta-doped ZrO_2 exhibits high dielectric constant without any previous high voltage sweep, retains its capacitance more than 30 minutes at elevated temperature of 75 degrees Celsius and endures more than 10^{12} cycles at 100 kHz cycling showing both robustness and reliability. Strain-mediated dielectric enhancement therefore provides a materials-design framework for achieving morphotropic-like dielectric responses in scalable fluorite thin films.

11:00am **AM+EM+TF-ThM-13 Ferroelectricity in Atomic Layer Annealed Nitride-Based Thin Films**, ***Andrew Meng***, *Dominic Dalba, Dilan Gamachchi, Indeewari Karunarathne, Bipin Bhattarai,* University of Missouri-Columbia; *Xiaoman Zhang,* University of North Alabama; *Wangwang Xu,* Louisiana State University; *Somayeh Saadat Niavol,* University of Missouri-Columbia; *Dongmei Cao, W.J. Meng,* Louisiana State University **INVITED**

Wurtzite AlN-based materials exhibit strong ferroelectric behavior and offer potential for a wide memory window due to their high coercive field, making them promising candidates for non-volatile memory applications. Most studies on wurtzite AlN-based ferroelectrics focus on reactive sputtered thin films, which can suffer from deleterious effects of point defects caused by ion bombardment. Atomic Layer Deposition (ALD) can lead to films with fewer electronic defects at significantly lower growth temperatures. However, the crystallinity can be low even in plasma assisted processes. Nitrogen incorporation in ALD films also remains challenging. We demonstrate the growth of nanocrystalline wurtzite AlN thin films deposited by a three-step atomic layer annealing (ALA) process at 350°C consisting of trimethylaluminum and hydrazine precursor pulses and a nitrogen plasma annealing step. Interestingly, these films are ferroelectric without intentional extrinsic alloying, which we attribute to very high dielectric strength in excess of 10 MV cm^{-1}. The films exhibit a remanent polarization of ~ 35 μC cm^{-2}, and piezoresponse force microscopy experiments show phase and amplitude changes after DC poling consistent with ferroelectricity. We also demonstrate growth of AlBN using an ALA process. The ALA growth process for wurtzite AlN-based ferroelectric materials is potentially compatible with processing for silicon electronics and holds promise for next-generation semiconductor devices.

[1] **JVST Highlighted Talk**

11:30am **AM+EM+TF-ThM-15 Ferroelectric and Rf Electrical Properties of Peald Scaln Thin Films**, ***Virginia Wheeler***[1], *Vikrant Gokhale, Margo Staruch,* US Naval Research Laboratory*; Jaime Hart,* Nova Research Inc*; Jonathan Chin, Peter Litwin,* NRC, US Naval Research Laboratory*; Neeraj Nepal,* US Naval Research Laboratory*; Gilbert Rayner, Jr, Noel O'Toole,* Kurt J. Lesker Company*; Nicholas Strnad,* Army Research Laboratory

Scandium aluminum nitride (ScAlN) has emerged as a transformative material for advanced microelectronics, driven by its exceptional piezoelectric response and robust ferroelectric properties. While traditional deposition methods, such as magnetron sputtering and molecular beam epitaxy (MBE), have enabled its initial integration, these line-of-sight techniques inherently lack the conformality required for next-generation, three-dimensional device architectures. Plasma-enhanced atomic layer deposition (PEALD) addresses this critical manufacturing bottleneck by providing self-limiting, atomic-scale control over film thickness and composition, enabling the uniform coating of complex, high-aspect-ratio structures. These benefits make PEALD ScAlN appealing for advancements in RF filters, where sub-nanometer thickness uniformity directly dictates acoustic performance and high-frequency bandwidth, and high-density 3D non-volatile memory (NVM) architectures requiring high-quality ultrathin layers. Consequently, the development of PEALD processes for ScAlN represents a critical pathway toward achieving next-generation, scaled, high-performance communication and at the edge computing technologies.

Using the PEALD process developed by Rayner et. al [1], 20-60nm ScAlN films were deposited on 50nm NbN/sapphire and Pt/SiO_2/Si templates with both 18% and 30% Sc. XRD and TEM confirm that all films are texture, polycrystalline in nature with roughness directly related to increasing Sc concentration and thickness. The 18% films exhibit low leakage, very flat permittivity (ε_r ~12-14) from 20 Hz to 3GHz, and low dielectric loss. However, increasing the Sc content results in more leaky films with strong dispersive permittivity and high loss, remarkably similar to MBE ScAlN/NbN films with 32% Sc.

Additionally, the 18% ScAlN/NbN film is confirmed to be ferroelectric, with measurements at the highest electric field achieved showing a remanent polarization value of 80 μC/cm^2. The coercive field, E_C, is approximately 5.5 MV/cm at room temperature and 50 kHz, comparable to previous reports of this composition. Although the measured polarization is below anticipated values, this is attributed to instrument limitations that precluded the application of higher electric fields, resulting in a minor loop with full saturation anticipated at increased field strengths. In-depth analysis of ferroelectric properties as a function of Sc content and substrate will be presented.

[1] Rayner et al, *JVSTA* 43, 020401 (2025).

11:45am **AM+EM+TF-ThM-16 Atomic-Scale Structure and Domain Mapping in Hafnia-Based Ferroelectrics**, ***Ece Gunay***[2], Carnegie Mellon University*; Sebastian Calderon,* Carnegie Mellon University, USA*; Rintaro Maki, Yuichi Shimakawa,* Kyoto University, Japan*; Daisuke Kan,* Osaka University, Japan*; Elizabeth Dickey,* Carnegie Mellon University, USA

Growing-memory architectures in logic circuits offer a promising pathway to reduce the energy cost associated with dense computing. These emerging device concepts require the integration of scalable, CMOS-compatible ferroelectric materials. Hafnium oxide, widely used as a gate dielectric, is inherently CMOS-compatible, and its ferroelectricity can be activated through cation alloying (e.g. Si, Zr, La, Y, etc.), which stabilizes a metastable orthorhombic phase and introduces new functionalities.

Because ferroelectric domains evolve and interact under applied bias, understanding their atomic-scale structure is critical for guiding device design. In this work, we investigated $Hf_{0.93}Y_{0.07}O_2$ freestanding membranes with in-plane polarization, enabling direct, real-space mapping of ferroelectric domains. Y-alloyed membranes were fabricated on (100) $SrTiO_3$ using a $La_{0.67}Sr_{0.33}MnO_3$ buffer layer, which was selectively etched to produce freestanding membranes for atomic-scale characterization.

Aberration-corrected scanning transmission electron microscopy (STEM) with annular dark-field (ADF) imaging was used to resolve cation positions, revealing ferroelectric domains oriented along the [001], [010], and [100] directions of the orthorhombic phase, forming 90° domain walls. Differential phase contrast (DPC) imaging enabled direct imaging of the oxygen sublattice. Vector pair correlation function analysis was used to map structure and domains across more than ten regions.

The results revealed that tetragonal-like inclusions are consistently observed near grain boundaries and domain walls. These inclusions are only a few unit cells thick and preserve overall lattice parameters while exhibiting local in-plane distortions. Additionally, lattice defects are frequently observed near 90° domain walls, likely accommodating strain arising from domain misorientation.

Overall, this study provides atomic-scale insight into the structural heterogeneity and domain behavior of hafnia-based ferroelectrics, offering guidance for optimizing their functional properties in next-generation electronic devices.

This material was based upon work supported by the Center for 3D Ferroelectric Microelectronics Manufacturing (3DFeM2), an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences Energy Frontier Research Centers program under Award Number DE-SC0021118.

12:00pm **AM+EM+TF-ThM-17 Correlating Biaxial Stress to Metastable Phases in $Hf_{0.5}Zr_{0.5}O_2$ on Substrates with Different Coefficients of Thermal Expansion**, ***Nooreen Qureshi***, University of Virginia, USA*; Luke Johnson,* Purdue University, USA*; Jordan Hachtel,* Oak Ridge National Laboratory, USA*; Jon Ihlefeld, Kory Burns,* University of Virginia, USA

Zirconium-substituted HfO_2 (HZO) has emerged as a leading candidate for next-generation ferroelectric memory and logic devices due to its Complementary Metal-Oxide-Semiconductor (CMOS) compatibility, scalability, and demonstrated ferroelectricity at thicknesses approaching 1 nm. Among the competing factors governing the ferroelectric phase stability in HZO including dopant density, oxygen vacancy concentration, electrode capping, and grain size; mechanical biaxial stress remains comparatively underexplored yet critically influential.

In this work, HZO thin films were deposited by Atomic Layer Deposition (ALD) on substrates with systematically varied coefficients of thermal expansion (CTE) to controllably tune the biaxial strain state. Macroscopic characterization via X-Ray Diffraction (XRD) and Infrared Spectroscopy (IR) reveal strain-dependent peak shifts that directly reflect the biaxial stress imposed by the substrate. Electrical measurements demonstrate a clear strain-dependent phase transition; films on low-CTE substrates exhibit ferroelectric polarization-voltage responses, while high-CTE substrates drive an antiferroelectric-like response, implicating strain as a tunable lever for phase stabilization.

To connect macroscopic behavior to local structural heterogeneities, scanning nanodiffraction 4D-STEM (Scanning Transmission Electron Microscopy) was employed to map lattice strain at the nanoscale. The correlation between local strain fields and bulk electrical response provides mechanistic insight into how nanoscale inhomogeneities propagate to device-level performance. This multi-scale characterization framework offers a comprehensive approach for engineering phase stability in fluorite-structure ferroelectrics.

[1] **JVST Highlighted Talk**
[2] **JVST Highlighted Talk**

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