PCSI
Room Ballroom South - Session PCSI-WeA2
Oxides III

Moderator: Alessandro Mazza, Los Alamos National Laboratory

4:30pm **PCSI-WeA2-31 Epitaxial Engineering of Emergent Phenomena in Tantalate Perovskites**, ***Kaveh Ahadi,*** Ohio State University **INVITED**

Epitaxial tuning knobs, including epitaxial strain, could serve as a powerful parameter that significantly alter the lattice symmetries, affect phase stability, and reshape the energy landscape. In this presentation I will discuss the epitaxial tuning of tantalate perovskites ($KTaO_3$ and $EuTa_2O_6$) grown using a sub-oxide molecular beam epitaxy method, which we recently developed for tantalates [1]. Next, I will talk about the effect of epitaxial tuning knobs, such as epitaxial strain, on the lattice and electronic structures. Here, I will discuss that$KTaO_3$, a cubic perovskite, can be epitaxially strained into a highly tunable ferroelectric. $KTaO_3$ films, grown commensurate to $SrTiO_3$ (001), experience an in-planecompressive strain of -2.1 % that transforms the otherwise cubic structure into atetragonal polar phase with a transition temperature of 475 K, consistent with ourphase-field calculations. The Curie temperature and the spontaneouselectric polarization are systematically controlled with epitaxial strain. Scanning transmission electron microscopy reveals cooperative polar displacements of thepotassium columns with respect to neighboring tantalum columns at room temperature. Optical second harmonic anisotropic rotation results are described by a tetragonal polar point group (*4mm*), indicating emergence of a global polar ground state.

Finally, I discuss our recent results on epitaxial control of ordering in fractionally occupied double perovskite, $EuTa_2O_6$ [2]. The intrinsic crystal anisotropy of $EuTa_2O_6$ plays a pivotal role, underscoring how targeted structural modifications can facilitate the emergence of novel quantum states. The crystal and electronic structures of $EuTa_2O_6$ are investigated. X-ray diffraction and electron microscopy reveal the layered *A*-site ordering. Angle-resolved photoemission spectroscopy, along with density functional theory calculations, provide direct insight into the electronic structure, unveiling the potential for engineered confined states within bulk materials. These findings highlight $EuTa_2O_6$ as a platform for studying 2D-like electronic phenomena in a 3D context, paving the way for novel device architectures.

[1] T. Schwaigert, S. Salmani-Rezaie, M. R Barone, H. Paik, E. Ray, M. D Williams, D. A Muller, D. G Schlom, K. Ahadi,Journal of Vacuum Science & Technology A **2**, 41(2023).

[2] T. Schwaigert, A. Barooni, B. Gregory, P. Malinowski, A. Tenneti, S. Hasko, B. Palazzolo, J. W Hodgson, B. Faeth, P. M Woodward, K. M Shen, A. Singer, M. Ghazisaeidi, S. Salmani-Rezaie, D. G Schlom, K. Ahadi, Advanced Functional Materials e13656 (2025)

Author for correspondence: Ahadi.4@osu.edu

5:10pm **PCSI-WeA2-39 Pockels Effect in Single-Domain a-Oriented BaTiO3 on Vicinal Si (001)**, ***Jason Tischler,*** University of Texas at Austin*; Agham Posadas,* La Luce Cristallina*; David Smith,* Arizona State University*; Kamyar Barakati, Kalinin Sergei,* University of Tennessee Knoxville*; Alexander Demkov,* University of Texas at Austin

Silicon (Si) integrated photonics is an emerging technology developed for the communications industry with emerging applications in quantum and neuromorphic computing [2, 3]. These devices provide advantages in data transmission rates by modulating optical signals in Si waveguides. However, Si by itself is a poor modulator of light as it has no intrinsic electro-optic effect. Barium titanate (BTO) demonstrates a large electro-optic response known as the Pockels effect due to it being a non-centrosymmetric medium. The Pockels effect is the change in refractive index of the material due to an external electric field. This response is described by a tensor and the largest component for BTO is r_{42}, which is reported to be 1,300 pm/V in bulk [4], and 923 pm/V in thin films [5,6]. To access this large value in a waveguide phase-shifter, one uses thin BTO films with the so-called a-axis orientation [7]. When BTO is epitaxially grown on Si (001) in the a-axis orientation, meaning the long c-axis is in-plane, two orthogonal in-plane crystallographic domains form. This creates problems in the form of Rayleigh scattering at the domain boundaries and not being able to fully utilize the high r_{42} coefficient.

In this talk we demonstrate the use of a 4° miscut vicinal Si (001) substrate to stabilize a single in-plane orientation growth of a-axis BTO on strontium titanate (STO) buffered Si by molecular beam epitaxy (MBE). In this study, utilizing x-ray diffraction (XRD) techniques and scanning transmission electron microscopy (STEM), we detail the crystalline microstructure confirming a single in-plane BTO orientation; The ferroelectric domain structure is characterized using piezo-force microscopy (PFM) and the electro-optic response is probed via transmission geometry Pockels measurements.

5:15pm **PCSI-WeA2-40 Epitaxial Single Crystal MgO Buffers on Si (100)**, ***Pablo Espinosa Argaiz****, Alexander Demkov,* University of Texas at Austin

The integration of functional metal oxides with silicon has been a persistent challenge due to the thermodynamic instability of their resulting interfaces [1]. Crystalline oxide buffer layers have been used as a solution by providing a thermodynamically stable transition layer. Magnesium oxide (MgO) buffers have been shown to work as effective pseudo-substrates for metal oxide epitaxy [2, 3], and its deposition on silicon has been shown to be stable [4].

In this talk we report an extensive study of high-quality MgO buffer layers using molecular beam epitaxy (MBE) on Si(100) substrates. The buffers were grown through electron-beam evaporation of single crystal MgO, which was deposited at 300°C under an oxygen partial pressure of $3.7x10^{-5}$ Torr. The reflection high-energy electron diffraction (RHEED) pattern (Figure 1.a) reveals a modulated pattern along the MgO[100] direction, indicating the beginning of small island formation and surface faceting. The thickness dependence on the films' crystallinity was studied through the FWHM of the (200) MgO Bragg peak rocking curve as seen in Figure 1.b. The trend suggests an improvement in the crystallinity as the layers become thicker. Figure 1.c shows the scanning transmission electron microscopy (STEM) image of a cross section of the MgO/Si interface, revealing an in-plane epitaxial relationship of MgO <100> || Si <100> with a 4:3 coincident site arrangement between the MgO and Si conventional unit cells. The figure also reveals the presence of tilted MgO grains and small-angle grain boundaries. The interface layer observed in Figure 1.c was investigated using x-ray photoelectron spectroscopy (XPS), suggesting the formation of Mg-O-Si bonds at the interface. Additionally, the measured band alignment at the MgO/Si interface and its relation to first principles calculations of various interface models will be discussed.

[1] K. J. Hubbard, D. G. Schlom, J. Mater. Res. 11, 2757 (1996).[2] K. Nashimoto, D. Fork, T. H. Geballe, Appl. Phys. Lett. 60, 1199 (1992).[3] D. K Fork, F. A. Ponce, J. C. Tramontana, T. H. Geballe, Appl. Phys. Lett. 58, 2294 (1991).[4] T. Abukawa, S. Sato, Y. Matsuoka, Surface Science 604, 1624 (2010).

5:20pm **PCSI-WeA2-41 An Operando Testbed for Emerging Analog Memories (OpTEAM)**, ***Alec Talin,*** Sandia National Lab

The rapidly growing amount of energy consumed in training and utilizing artificial intelligence such as the large language models (LLMs) has motivated an intense search for new computing hardware and algorithms to improve energy efficiency[1]. In-memory computing schemes based on emerging analog non-volatile analog memory (NVM) device concepts has the potential to dramatically lower the energy used for data movement in current digital architectures. However, despite numerous demonstrations of promising device characteristics for NVMs based on various mechanisms (e.g. filamentary, phase change, magnetic, etc.) the practical implementation of these approaches remains uncertain. This is due, in part, to non-ideal device characteristics such as non-linearity and excessive noise at high resistances, inadequate endurance and low retention. Some of these issues such as noise and non-linearity can be addressed using for example denoising procedures or various algorithms, but the increased latency and/or energy costs substantially lessen the appeal of the emerging NVM solution. Another challenge is the lack of well-defined fabrication processes, predictable and controllable characteristics and scalable, validated compact models for the design of arbitrary circuits and networks. The approach of fabrication and test of small batches of isolated devices often leads to poor reproducibility and is inadequate for the development of compact model needed for the design of functional networks. In this work, we demonstrate an operando testbed for emerging analog memory development (OpTEAM) (Fig. 1) designed for testing small networks (up to 16 devices) of NVMs without the need for integration with CMOS peripheral input/output circuitry or access devices. Additionally, the OpTEAM is designed to facilitate the correlation of electrical tests such as training or inference with other physical characteristics such as thermal or optical/spectroscopic measurements. As 'exemplar' devices we use TaO_x resistive memory and WO_x based electrochemical memory (WO_x-ECRAM). Taking advantage of the electrochromic quality of WO_x allows us to

correlate programming of the memory elements with the spatial distribution of the oxygen vacancy dopants.

[1] A. A. Talin and B. Yildiz, Introduction: Neuromorphic Materials, Chem. Rev 125, 4765, 2025.

+Author for correspondence: aatalin@sandia.gov

5:25pm **PCSI-WeA2-42 High-Temperature Resilient Neuromorphic Device based on Optically Configured Monolayer MoS2 for Cognitive Computing**, ***Pukhraj Prajapat****,*** *Govind Gupta,* National Physical Laboratory, India

High-temperature neuromorphic devices are becoming more and more essential as technology progresses to support space exploration and survive extreme conditions such as those found in factories. To overcome this need, the researchers are devising technologies that imitate human brain structure and operation. In this work, we present a scalable neuromorphic device based on a monolayer of MoS_2, that demonstrates operation at 100°C. The device portrays excellent electrical performances mostly due to the great thermal stability of monolayer MoS_2 and its mechanical flexibility. Among these performances are low power consumption, fast switching, high resistance ratio, low switching voltage, and long stable endurance (~10^3 cycles). Besides, the device mimics neuromorphic behavior by embedding the synaptic plasticity that is the major functional property of biological neural networks, thus allowing advanced cognitive computing in extreme environments. This is the first step toward a combination of materials science and neuromorphic computing, and it clears the way for smart resilient electronics that could survive in a variety of harsh conditions. This research is targeting a major change in the area of high-temperature electronics, and this progress is paving the way for obtaining future high-performance electronics that can meet the needs of modern technology.

5:30pm **PCSI-WeA2-43 Understanding Dielectric Breakdown Using EDMR and NZFMR**, ***Colin McKay****,*** Sandia National Laboratories*; George Bodenschatz, Kaila Burges, Elijah Allridge, Michael Elko, Patrick Lenahan,* Penn State University*; David Hughart, Gaddi Haase,* Sandia National Laboratories

Time dependent dielectric breakdown (TDDB) is a fundamental problem in solid state electronics which is still not fully understood. Different models in the literature provide very different expected lifetimes. A deeper understanding of the physical mechanisms of TDDB can be gained from using electrically detected magnetic resonance (EDMR) and near zero field magnetoresistance (NZFMR). This abstract shows data from such a study. We report a fundamental advance in our understanding of TDDB in SiO_2 and the first direct observation of the generation of a specific point defect, the E' center, due high field gate stress using EDMR, NZFMR, and other techniques at room temperature. EDMR and NZFMR are spectroscopic techniques sensitive only to electrically active defects.

In this study, gate oxides in large arrays of silicon on insulator (SOI) n-MOSFETs were subjected to high electric field stress at 7.5V. Damage caused by the stress was characterized using the Fitzgerald-Grove gated diode method, capacitance vs voltage (CV), EDMR, and NZFMR measurements. The gate oxides were 7 nm thick and the gate areas of the transistor arrays were between 5,000 μm^2 and 50,000 μm^2.The early increase in peak DCIV current indicates that the first stage of damage is characterized by interface state generation, specifically Pb centers, with no appreciable increase in bulk oxide defects. The generation of interface states is accompanied by the redistribution of hydrogen away from recombination centers shown by the NZFMR results. The interface state density eventually starts to saturate, followed by an increase in bulk oxide defects, specifically E' centers, represented by a shift in the voltage of the DCIV peak current. The Pb and E' centers were identified via their EDMR signals. The E' spectrum only appears after long stress durations. This new understanding of the different stages of damage provides a fundamental insight into the physics of damage mechanisms during the leadup to TDDB.

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5:35pm **PCSI-WeA2-44 Interfacial Polarization in Polymer-Based Dielectric Composites with 2d Nanomaterials**, *So-Yeon Jun,* ***SeGi Yu****,* Hankuk University of Foreign Studies, Republic of Korea

The dielectric behavior of polymer-based composite films was systematically investigated with a particular focus on the interfacial polarization phenomena arising from the incorporation of conducting two-dimensional (2D) nanoplatelets. Ferroelectric $BaTiO_3$ (BTO) nanoparticles were employed as a primary filler; while MXene, graphene oxide (GO), and reduced GO (rGO) served as co-fillers to modulate the interfacial effects. Cyanoethyl pullulan (CEP) was used as a polymer matrix due to its high dielectric behavior among polymers. A reference sample, containing BTO nanoparticle fillers only in a CEP matrix, was also prepared to isolate the contribution of the 2D nanomaterials, which resulted in four dielectric composite samples. MXene platelets were delaminated in dimethyl sulfoxide (DMSO) via ultrasonication for 2 hours, and rGO was chemically reduced from GO using hydrazine over 8 hours—both conditions optimized in prior studies [1,2] to enhance dielectric performance. All components were dispersed in dimethylformamide (DMF) and spin-coated onto ITO substrates to form uniform dielectric films.

The incorporation of conducting 2D nanoplatelets significantly enhanced the dielectric constant of the composites. The highest value was achieved with the MXene-incorporated film (~200 at 1 kHz), followed by GO films (~130) and rGO films (~120), all substantially higher than the reference film containing BTO only (~90). However, this enhancement was accompanied by an undesirable increase in the dielectric loss (tan δ) due to percolative linkage of fillers. The loss increased, from 0.048 for the reference sample, to 0.053 for GO, 0.10 for MXene, and 0.22 for rGO, respectively.The observed dielectric enhancement is attributed to pronounced interfacial polarization at the filler-matrix boundaries, facilitated by the high aspect ratio and conductivity of the 2D nanoplatelets. Among them, MXene demonstrated superior interfacial coupling due to its metallic conductivity, leading to more effective charge accumulation at interfaces. In addition, MXene can successfully suppress the increase in the dielectric loss which is difficult to control for nanomaterial incorporation within a polymer matrix. Raman spectroscopy and X-ray photoemission spectroscopy (XPS) analyses corroborated the structural and electronic characteristics responsible for these effects. These findings demonstrate the critical role of interfacial polarization in tailoring the dielectric properties of polymer-based composites and suggest that MXene-based systems hold promise for next-generation electronic and energy storage applications, where high permittivity and controlled loss are essential.

5:40pm **PCSI-WeA2-45 Switchable Electron-Phonon Scattering Strength in Monolayer Hexagonal Boron Nitride**, *Alv Johan Skarpeid,* University of Oslo, Norway*; Noah Joseph Hourighan,* Graz University of Technology, Austria*; Richard Justin Schenk,* University of Oslo, Norway*;* ***Håkon Ivarssønn Røst****,* University of Bergen, Norway*; Giovanni Di Santo, Luca Petaccia,* Elettra-Sincrotrone Trieste, Italy*; Bodil Holst,* University of Bergen, Norway*; Anton Tamtögl,* Graz University of Technology, Austria*; Justin William Wells,* University of Oslo, Norway

In the past decade, the layered compound hexagonal boron nitride (hBN) has drawn considerable attention due to its compatibility with various low-dimensional van der Waals (vdW) materials [1]. While hBN resembles graphene in lateral size, crystalline structure, and Debye frequency, its two distinct sub-lattices give rise to a significant energy band gap between the valence and conduction bands [2]. Recently, it was predicted that hBN should host strong electron-phonon coupling (EPC) in its electronic π- and σ-bands [3], reminiscent of the reported (and debated) interactions in the graphene σ-bands [4]. Since then, we have confirmed this EPC from observable energy renormalizations in the hBN band structure [5].

We will showcase the electron-phonon coupling (EPC) in hBN, highlighting how changing the substrate interaction, e.g., by adatom intercalation, can influence coupling strength (see Fig. 1). By combining angle-resolved photoemission and neutral helium atom scattering, we will demonstrate how these techniques together help demystify the scattering modes involved in electron-hole recombination. We will also mention the broader implications of EPC in materials with finite electronic band gaps.

[1] Y. Lui et al., *Nat Rev. Mater*. **1**, pp. 1-17 (2016).

[2] J. Robertson, *Phys. Rev. B* **29**, p. 2131 (1984).

[3] E. Thingstad et al., *Phys. Rev. B* **101**, p. 214513 (2020).

[4] F. Mazzola et al., *Phys. Rev. B* **95**, p. 075430 (2017).

[5] H. I. Røst et al., *Nano Lett*. **23**, pp. 7539-7545 (2023).

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